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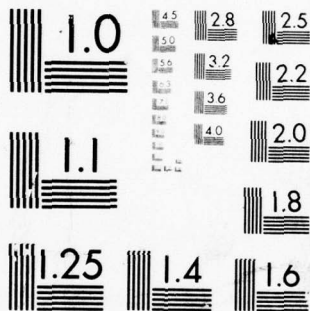
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CONTRACTOR REPORT  
ARCSL-CR-77050

A STUDY OF AEROSOL ASSESSMENT INSTRUMENTATION

Final Report

by

Madhav B. Ranade

September 1977

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Under this program, an up-to-date survey of commercial instruments available for measurement of aerosols was conducted. Aerosol properties of interest were size, concentration, stability, and composition. Aerosol instruments were classified according to their measurement principles and capabilities. An analysis of cost effectiveness for a specific application was performed after identifying the information required and instrument capabilities.			

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### Preface

The work described in this report was authorized under Project No. DAAAL5-76-C-0122, "A Study of Aerosol Assessment Instrumentation". This work was started in June 1976 and completed in October 1976.

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## A STUDY OF AEROSOL ASSESSMENT INSTRUMENTATION

### 1. INTRODUCTION

The Munitions Application Branch is responsible for the study and determination of the dissemination characteristics of chemical agents. This responsibility includes the determination of particle size distribution and concentration of aerosols in closed test chambers.

The objective of the present contract was to determine the most reliable, accurate, and cost effective instrumentation for use by the Munitions Applications Branch in the assessment of experimental aerosols. Recent developments in aerosol science and technology have led to the development of several new techniques. Comparative evaluation of these is needed so that the Munitions Applications Branch may keep abreast of the latest instruments.

An aerosol consists of an assembly of discrete particles dispersed in a gaseous medium. Properties of an aerosol system depend on the properties of the individual particles, their interactions with the medium, and the interactions between the individual particles. The particle size represents a linear dimension assigned to a particle by methods which typically measure quantities related to either the number, linear size, surface area or the volume (or mass) of the particles. Even for ideally shaped spherical particles, conversion of one measured quantity into another may cause significant errors.

The reliability of an assessment method for an aerosol under study is related to the aerosol property under study. The chosen method of measurement should be such that the quantity or dimension measured is of the same form as the property to be studied. For example, the aerodynamic size of the particles is probably the most meaningful quantity in the study of aerosol particle motion. Use of a method based on the inertial classification is indicated in this example.

Instruments for aerosol assessment at various levels of sophistication are available commercially. Some of these have been around for a long time and have been tested in various applications so that their capabilities are well documented. The new generation of instruments, due to the progress in technology, have extended assessment capabilities to high levels of sophistication in a relatively short time and their performance has yet to be fully documented. Judgment on applicability of these instruments should be based on a careful consideration of the measurement principle in relation to the desired application.



In this report, we have compiled documented information on the instruments available commercially, making use of the manufacturer's literature and published literature. We have also discussed the measurement principles and their applicability to the needs of the Munitions Application Branch.

The Munitions Applications Branch is primarily interested in two aerosol systems to be studied in a large chamber. The first system consists of particles in 0.1-1.0  $\mu\text{m}$  nominal size range and concentrations of up to  $10^6$  particles/ $\text{cm}^3$ . The particles are mainly liquid and may be hygroscopic. The second system consists of particles in 1-100  $\mu\text{m}$  nominal size range and concentrations of up to  $10^6$  particles/ $\text{cm}^3$ .

We have established several combinations of instruments and auxiliary equipment and an analysis of their benefit and cost is also presented. Several factors were considered as having significant effects on the performance and were weighed depending on the nature and magnitude of their effect. Similarly, the desired features required of the instruments for a given application were also weighed according to their importance. The costs of several systems were compared to the achievable benefits.

## 2. INSTRUMENTS FOR AEROSOL ASSESSMENT

Instruments for the measurement of aerosols are based on sensing properties of aerosols which are related to the number of particles, linear size, surface and the volume (or mass) of the particles. Consequently, they suffer from limitations of effectiveness over ranges in which the properties are significant or measurable.

An aerosol system is characterized by the size distribution and the concentration of the particles. Size distribution of the aerosol particles is expressed in several manners. Most common of which are the histograms or percent distributed in a given size range. The distribution may be based on count or mass or volume of the particles. In Table 1, several methods of sizing aerosol particles, the characteristic measured quantities, bases, and factors to be considered for reliability are shown. Depending on their levels of sophistication, all aerosol instruments have one or more of the following functions: (1) identification, (2) classification, and (3) measurement. Some of the instruments are based on processing a signal from individual particles one at a time, or a collective signal from several particles in the sensing.

At the first level of sophistication, the aerosol photometers measure the light scattered by an assembly of aerosol particles in a sensing zone. The signal intensity is related to the loading or the concentration of the particles in the

Table 1

## METHODS FOR AEROSOL ASSESSMENT

Identification Method	Classification Principle	Sensed Property	Characteristic Quantity	Basis	Reliability Considerations
Scattered Light	--	Light Intensity	Scattering Cross-section	Number	Refractive Index Shape
Mass	Inertia	$\propto$ Mass	Aerodynamic Diameter	Number Mass	Shape, Density
Mass	Electrical	$\propto$ Mass	Aerodynamic Size	Mass	Shape, Density Charge
Scattered Light	--	Light Intensity	Scattering Cross-section	Number	Refractive Index Shape
Light Extinction	--	Light Intensity	Extinction Cross-section	Number	Refractive Index Shape
Image	--	Projected Area	Area	Number	Shape
Electrical Charge	Mobility & Aerodynamic	Current	Mobility	Number	Shape



aerosol over a wide range of particle sizes. No information of the size distribution of the particles is obtained. Condensation nuclei counters incorporate a controlled growth of very small particles ( $2.5 \times 10^{-3}$  -  $1 \mu\text{m}$ ) followed by a scattering or extinction measurement to determine concentration of submicron particles. In the quartz crystal monitors and  $\beta$ -attenuation monitors, the particles are deposited by inertial or other means and their mass is measured. A classification step before the deposition may be used to determine mass of aerosol particles in several size ranges. These instruments operate on the aerosol particle assembly as a whole. Electrical field classification followed by a charge measurement is also used to measure the size distribution of submicron aerosols.

Single particle analyzers process signature of individual particles in several classes which are dependent on the signal levels. In most devices, the particles are made to flow through the sensing volume one at a time.

In another type of counter, a two-dimensional image of the particle is produced and then electrically processed to analyze the image based on linear dimensions and projected areas of the particle images.

## 2.1 Instruments for Concentration Measurement

### 2.1.1 Aerosol Photometers

Forward light scattering photometers, such as the Sinclair-Phoenix, are useful for monitoring concentration of particles over wide ranges of size and concentrations. A schematic diagram of the Sinclair-Phoenix is shown in Figure 1. A tungsten filament lamp is used as the light source. A series of condensing lenses and a central dark spot produce an axially symmetric hollow cone of illumination focused on the aerosol stream. Light scattered in the forward direction in  $5^\circ$ - $30^\circ$  zone is collected by a lens and transmitted to a photomultiplier tube. The signal intensity is dependent on particle size, refractive index, and number concentration. The integral signal may be calibrated against mass concentration of the aerosol particles obtained by determining the mass collected on a filter. Any changes in particle characteristics and concentration will alter the calibration. For this reason, this instrument is used only in limited applications such as filter testing where the test aerosol parameters may be kept constant by using a narrow size distribution. In another application, the signal is used to relate the atmospheric visibility reduction due to aerosols.

In the integrating nephelometer shown schematically in Figure 2, light scattered over almost the entire  $360^\circ$  is sensed to achieve increased reliability. As in the Sinclair-Phoenix,

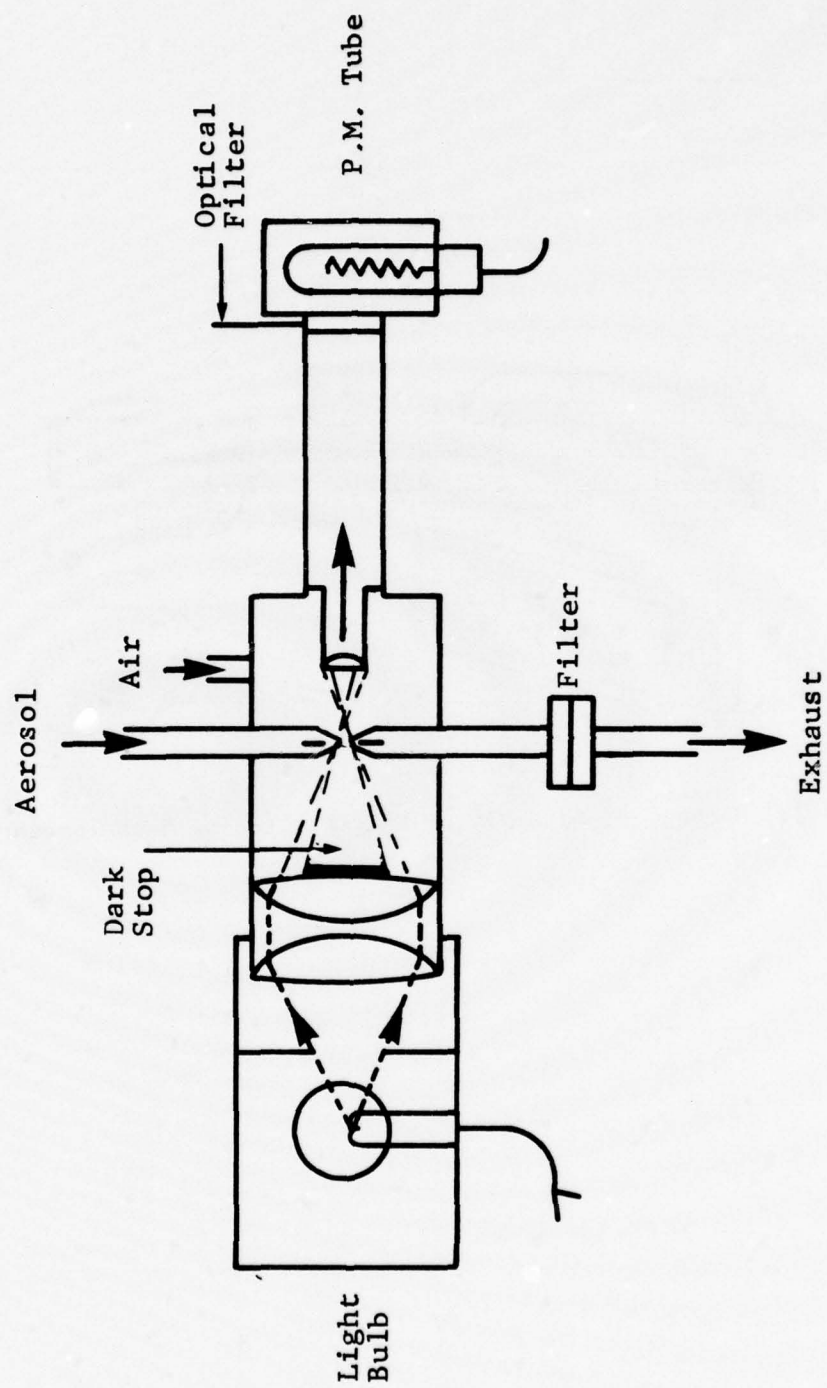


Figure 1. Forward Light Scattering Photometer

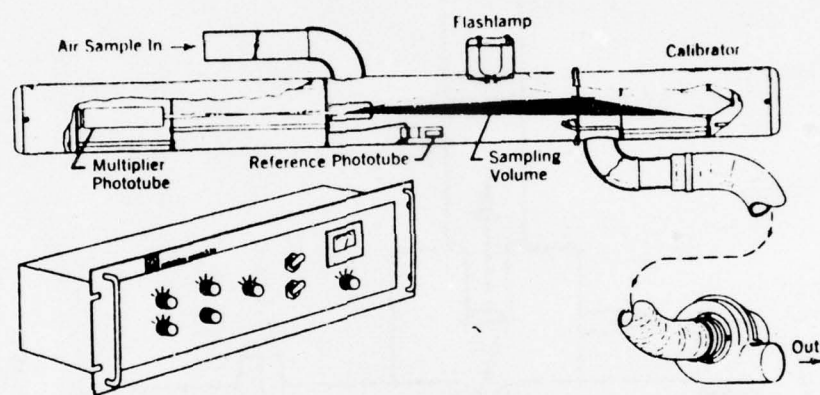


Figure 2: Mechanical Assembly of MRI Integrating Nephelometer

this instrument is useful for determination of visual quality of atmosphere and mass loading of a stable aerosol after calibration.

A list of commercially available instruments of these types and their manufacturer is given in Table 2. Specification sheets and brochures were supplied separately to the Project Officer.

#### 2.1.2 Condensation Nuclei Counters

Nolan and Pollack [1] developed the first apparatus for determining concentrations of Aitken nuclei. An aerosol in atmosphere saturated with water vapor is adiabatically expanded to produce instantaneous supersaturation of up to 400%. Water is condensed on the nuclei to form a fog. Attenuation of light by this fog is measured and calibrated against the number of nuclei.

An inexpensive manually operated instrument operating on this principle is manufactured by Gardner Associates. Automated versions of the instrument are made by Environment-One Inc. and General Electric Corporation. A summary is presented in Table 3. Schematic diagrams of these instruments are shown in Figures 3, 4, and 5.

The condensation nuclei counter is capable of detecting particles from  $2.5 \times 10^{-3}$  to  $1 \mu\text{m}$  in size. The lower limit of detectability is determined by the adiabatic expansion ratio.

O'Connor [2] reported on calibration of a manual and an automatic commercial counter. The primary calibration of the ambient test aerosol was performed using an original Nolan-Pollack counter. Good agreement between the instruments was observed as shown in Figures 6 and 7.

Due to variation in nucleating efficiency of particles of different compositions, an aerosol of known size and composition should be used for the calibration rather than the heterogeneous mixture of ambient aerosols. Liu and co-workers [3,4] at the University of Minnesota used a monodisperse aerosol for more rigorous calibration. Their results indicate that the automatic counters predict the concentration low at high concentration levels by a factor of almost 6. Their results are shown in Figures 8 and 9.

The nucleating efficiency of the particles is affected by several factors such as wettability by water and hygroscopicity. In our experience, hygroscopic aerosols grow large enough without the adiabatic expansion so that at high concentrations ( $>15$  particles/cm<sup>3</sup>), reference signal is large enough to prevent measurement of the fog resulting from the adiabatic expansion.



Table 2

## A LIST OF COMMERCIAL AEROSOL PHOTOMETERS

Manufacturer	Model	Particle Size Range (sensitive range), $\mu\text{m}$	Concentration Range
Phoenix-Precision Gardiner, N.Y.	Sinclair-Phoenix JM 4000	>0.05-40	$1-100,000 \text{ particles/cm}^3$ (order of range)
Sartorius	S	>0.05-40	$5 \times 10^2 -$ $10^8 \text{ particles/cm}^3$ (0.3 $\mu\text{m}$ dia.)
Frontier		>0.05-40	$1-100,000 \text{ particles/cm}^3$ (order of range)
Meteorology Research, Inc.	Integrating Nephelometer	>0.01	$0-3,800 \mu\text{g/m}^3$
Climet	CL-260	0.01-10	$0-100,000 \mu\text{gm/m}^3$ (0-3 $\mu\text{m}$ dia)
Sigrist Photometer Ltd.	TN	0.3-1,000	$0-1 \text{ gm/m}^3$

Table 3  
COMMERCIAL CONDENSATION NUCLEI COUNTERS

Manufacturer	Model	Particle Size Range $\mu\text{m}$ (sensitive range)	Concentration Range particles/cm <sup>3</sup>	Operative Mode
BGI, Inc. Waltham, Mass.	P	$1 \times 10^{-2} - 1$	10-500,000	Manual
Environment One Corp. Schenectady, N.Y.	Rich 100	$1.6 \times 10^{-3} - 1$	$300-10^7$	Automatic
Gardner Association Schenectady, N.Y.	CN	$2 \times 10^{-3} - 1$	$200-10^7$	Manual
General Electric Pittsfield, Mass	CNC-2	$1 \times 10^{-3} - 0.1$	$10-10^6$	Automatic

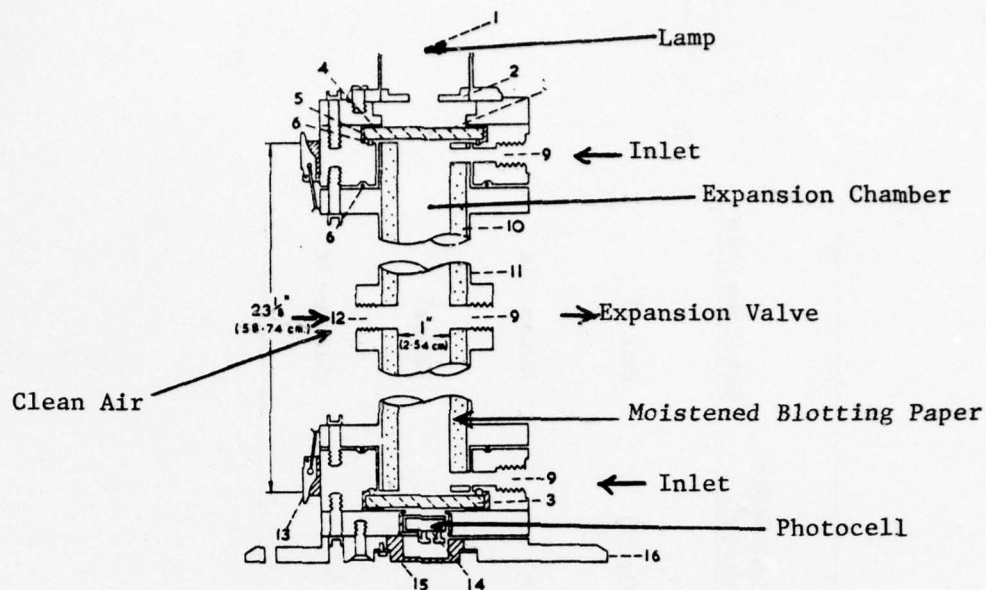


Figure 3: Diagram of Nolan Pollak nucleus counter.

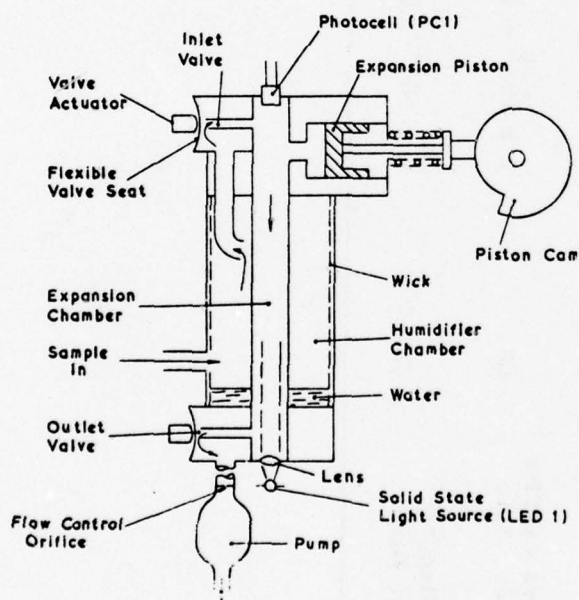


Figure 4: Schematic of 'Environment-One' portable nucleus counter.

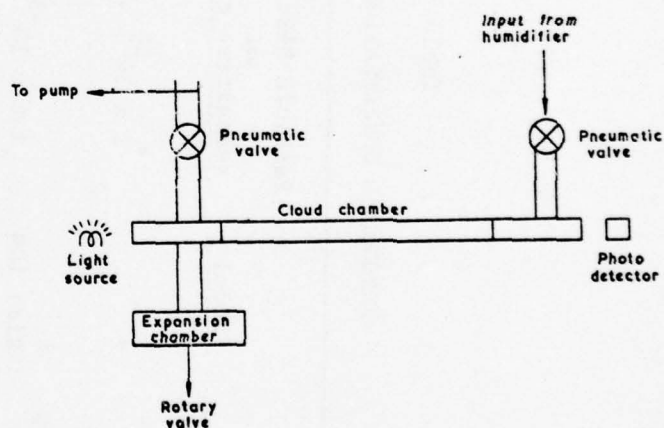


Figure 5: Schematic of 'Environment-One' automatic nucleus counter.

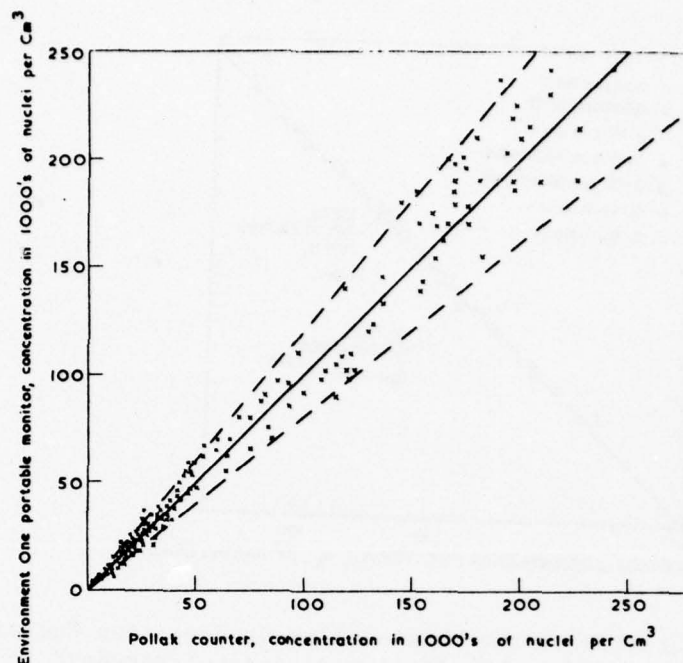


Figure 6: Concentrations of condensation nuclei as read by Environment-One Portable Monitor vs. those as read by a Pollak Counter.

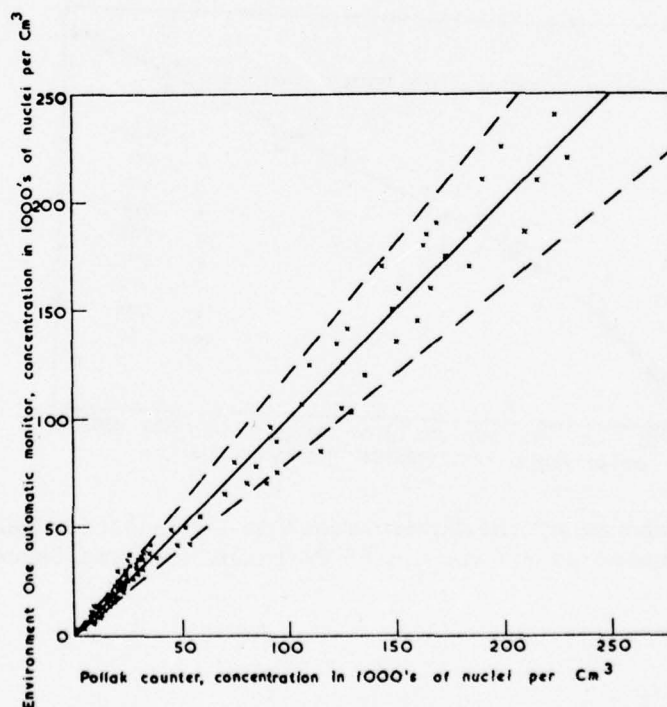


Figure 7: Concentrations of condensation nuclei as read by Environment-One Automatic Monitor vs. those as read by a Pollak Counter.



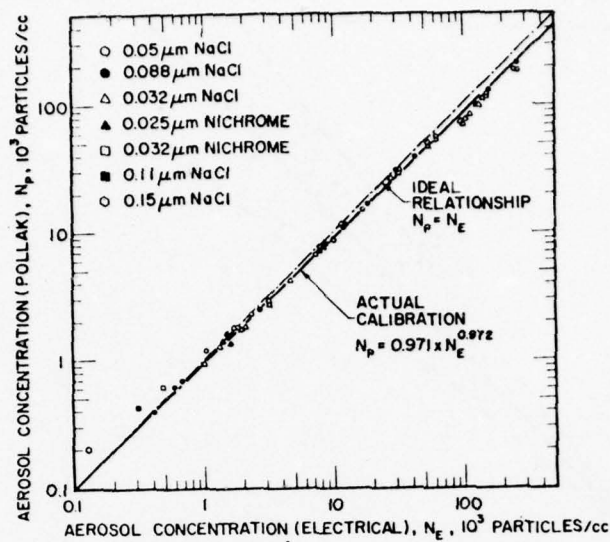


Figure 8: Calibration of the Nolan-Pollak Condensation Nuclei Counter Against the Electrical Aerosol Standard

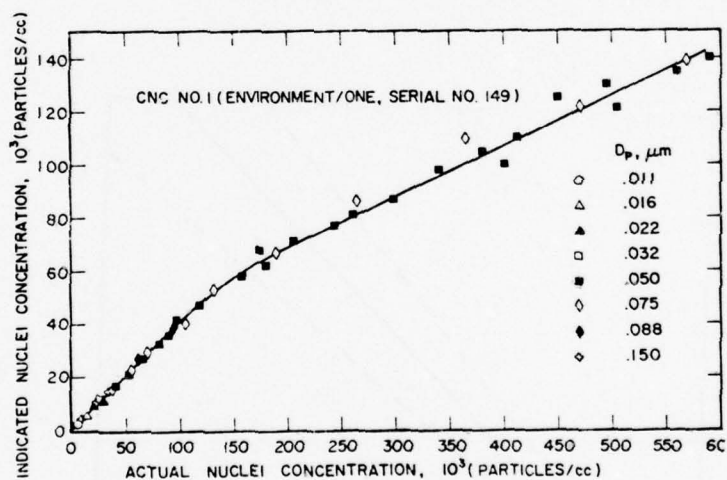


Figure 9: Response of the Environment/One Condensation Nuclei Counter as a Function of Particle Size and Concentration

### 2.1.3 Surface Ionization Monitor

When particles come into contact with a hot surface, they decompose. Atomic and molecular constituents of the particulate adsorb briefly on the surface. The constituent atoms having ionization potential comparable to the work function of the hot surface are released as positively charged ions. For submicron and micron sized particles, the ions are released as a burst which produces an electrical pulse in the detector. The pulses representing individual particles are analyzed by a pulse height analyzer or may be integrated to display a signal related to the assembly of particulates. A device called SIMP-1, based on this technique is manufactured by Extranuclear Laboratories, Inc., Pittsburgh, Pa.

This technique is responsive to a limited number of atomic and organic molecules. Among the elements that are detectable [5] are Li, Na, K, Rb, Cs, Al, Ba, Ca, Eu, In, La, Pr, Th, and Uranium.

A hot wire is used as a hot collector surface. Electrical potential difference between the collector wire and an ion collecting electrode causes the ions to migrate to the electrode.

Besides the limitations of its application to only a few elements, collection of the particles by the hot wire is questionable. Interception is the only mechanism used for collection and is not considered effective for particles below a few microns. In addition, the thermal gradient between the wire and the surrounding enclosure may repel submicron particles away from the wire by thermophoresis.

In conclusion, the instrument may be considered as having a rather limited, if any, quantitative usefulness.

### 2.1.4 Charge Transfer Particle Monitor

When dissimilar materials come into contact, charge transfer takes place due to the contact potential difference. This phenomena has been utilized in the particulate monitor manufactured by IKOR, Inc. [6]. If particles in a flowing stream are allowed to impact on an electrically isolated sensing zone continuously, charge transfer will take place resulting in a small current. The particles need not be charged before impaction. The current is processed electronically so that a voltage output proportional to the charge generated by the particle-sensor collisions is produced.

The instrument was originally developed for sampling of dry particulate from stacks. The detection capability is claimed over a 0.1-100  $\mu\text{m}$  range of particle sizes. The concentration range is 23 to  $23 \times 10^8 \mu\text{g}/\text{m}^3$ .

The Model 207 is a portable particle monitor. Aerosol sample is transported through a sampling probe to the sensor. Model 2710 is designed so that the sensor mounted on the tip of the probe may be positioned in an aerosol stream.

Although the charging of the particles is not necessary for the device to function, the level of the signal is affected by changes in charge inventory of the sampled aerosols. The signal magnitude is also dependent on the composition of the particulate material and its electrical insulation characteristics (conductor, semi-conductor or insulator). The reliability of the signal is dependent on the nature of the sensor surface. Wet or oily surface will affect the sensor electrical properties. Use is recommended only with dry material.

In general, the device is useful for aerosols of steady composition and size distribution. The signal must be calibrated against mass loading determined by another method such as gravimetric sampling on a filter.

#### 2.1.5 Beta Ray Attenuation

$\beta$  radiation passing through a substrate carrying a particle deposit is absorbed and intensity is attenuated as given by

$$\frac{N}{N_0} = e^{-\mu_m \delta} \quad (1)$$

where  $N$  and  $N_0$  are the  $\beta$  counts with and without particles,  $\mu_m$  is the characteristic absorption coefficient of the  $\beta$  absorbing substrate, and  $\delta$  is the average mass per unit area of the collected particulate layer. From equation 1, it follows that the mass " $m$ " of particles on a substrate is given by

$$m = \frac{A}{\mu_m} (\ln N_0 - \ln N) \quad (2)$$

where  $A$  is the effective deposit area.

GCA/Technology Division manufactures two types of  $\beta$ -ray attenuation particle monitors. RDM-101 utilizes an impactor to deposit the particles on a polyester disc. A carbon-14 source is used for  $\beta$  radiation. A Geiger Mueller detector is used to measure the  $\beta$  radiation. The schematic drawing of the instrument is shown in Figure 10. The second type of instrument utilizes a filter tape for sample collection. The sample is intermittently pulled through the filter for a pre-set time. Macias and Husar [7] point out that mass attenuation



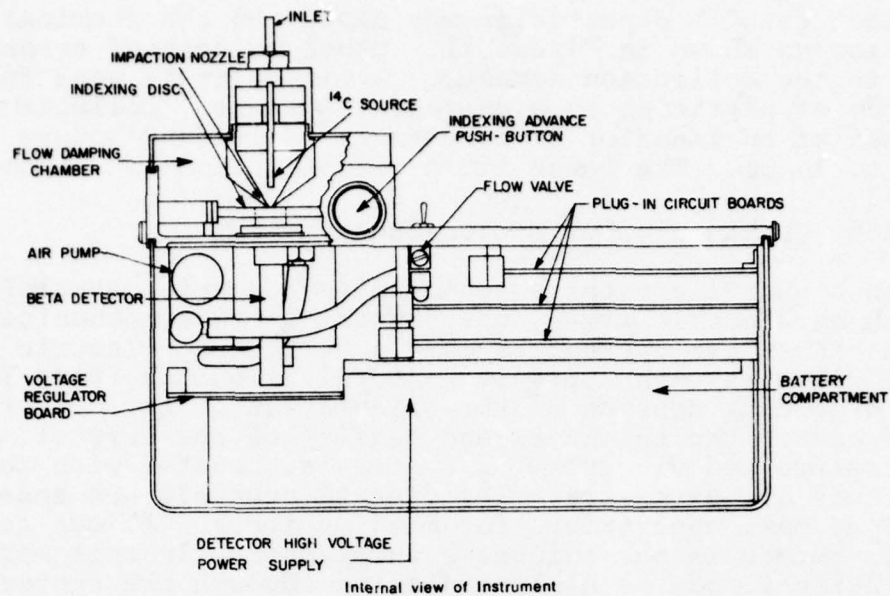


Figure 10: RDM-101 B-Attenuation Mass Monitor (GCA).

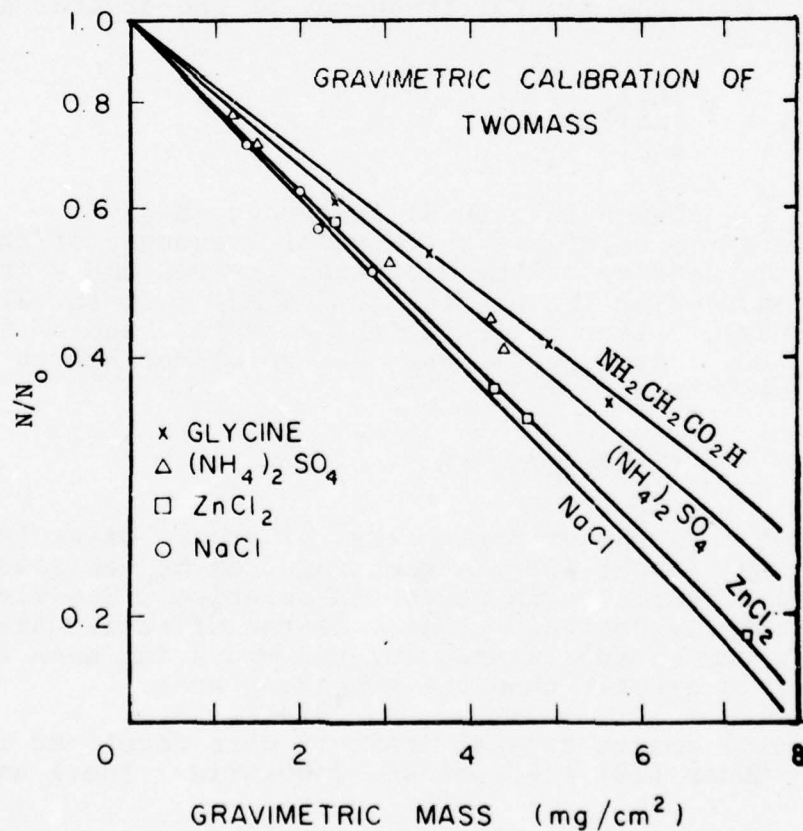


Figure 11: Effect of Chemical Composition on B-Attenuation.

coefficient for  $C^{14}$   $\beta$ -particles may depend on the chemical composition as shown in Figure 11. Other sources of errors are due to the collection schemes. An impactor is used for deposition of particles on a polyester surface. Collection efficiency of an impactor is inherently limited by bounce problem to 15  $\mu\text{m}$ . The lower limit of collection is 0.4  $\mu\text{m}$ .

#### 2.1.6 Quartz Crystal Microbalances (QCM)

When a quartz crystal or other piezoelectric material is placed in an electric field, the crystal becomes mechanically stressed. When the crystal is made a part on an electric circuit, it vibrates with a precise natural frequency [8]. The natural frequency depends on the orientation of the cut of the crystal, and on the thickness and density of the crystal. Due to temperature and vibratory parameters associated with the various cuts of quartz crystals, type AT crystals are generally accepted as most appropriate for mass monitors. AT-cut quartz crystals vibrate in the thickness shear mode. In this mode, the vibrational mode is a plane passing through the center of the crystal, between the flat surfaces of the plate-like crystal. Only the portion of the crystal between the electrodes (Figure 12) vibrates.

Particles are deposited on the crystal surface by several mechanisms. The presence of particles on the crystal surface causes a shift in the natural frequency of the crystal according to the relation

$$\Delta f = - \left( \frac{C_f}{A} \right) \Delta M \quad (3)$$

where  $f$  is the change in natural frequency,  $\text{MHz}$ ,  $C_f = f_0^2 / \rho N$  ( $\text{MHz}^2 \text{cm}^2 / \text{gm} \cdot \text{cm}$ )  $f_0$  is the natural frequency of the crystal,  $\rho$  is the density of the vibrating crystal and  $N$  is the frequency constant of the crystal (0.166  $\text{MHz} \cdot \text{cm}$  for AT-cut quartz crystal),  $A$  is the area of the substrate and  $\Delta M$  is the mass gained due to the particles. For an AT-cut quartz crystal equation 3 reduces to

$$\Delta f = - (2.27 f_0^2 / A) \Delta M \quad (4)$$

The area  $A$  is the vibrating crystal area. Daley [9] has shown that the crystal and the electrode can be designed to damp out non-linearities in  $\Delta f$  vs.  $\Delta M$  relation. The electrode area in a properly designed crystal is the effective area. The deposit area may be substituted for the vibrating area if the deposit area is greater than the vibrating area.

Commercial quartz crystal monitors were developed following the work of Chuan [10] (IBC Celesco Industries, Inc.) and Olin

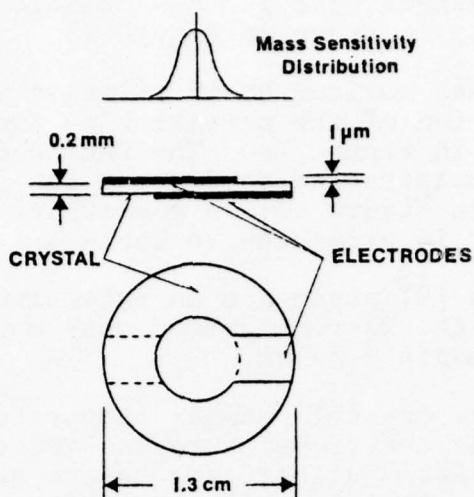


Figure 12: Sketch of Typical Crystal Showing Approximate Mass Sensitivity Distribution

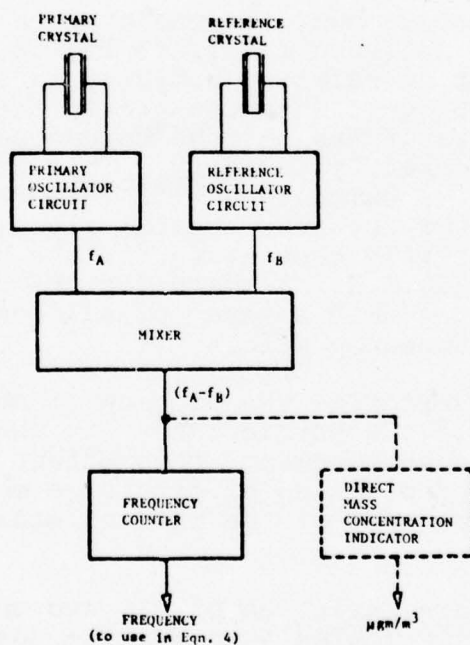


Figure 13: Piezoelectric Microbalance Electronic Block Diagram

and Sem [11] (Thermo-Systems, Inc.). A schematic diagram of the electronic circuitry is shown in Figure 13.

The IBC-Celesco mass monitor PM-40 utilizes an impactor arrangement for deposition of the particles on the crystal as shown schematically in Figure 14. The Thermo-Systems unit utilizes electrical precipitation to deposit the particles on the crystals as shown in Figure 15. A summary of the design and operating parameter is presented in Table 4.

Daley and Lundgren [9] performed an extensive evaluation of the IBC-Celesco and the Thermo-Systems mass monitors. Their findings are summarized below.

For both units, the crystal chamber temperature remained constant while the inlet air temperature was varied cyclically. The test crystal experienced higher temperature gradient than the reference crystals. The effective temperature coefficients based on the inlet temperature were  $0.9 \text{ Hz}/^{\circ}\text{C}$  for the TSI 3200 unit. A temperature change of  $1^{\circ}/\text{min}$  would be required to produce an error of  $5 \text{ } \mu\text{g}/\text{m}^3$ . Use of a metal tube in the sampling line, which would act as a heat sink, was found to reduce the temperature fluctuations.

Effect of the relative humidity was studied with and without the particulates. As shown in Figure 15, in absence of particulates the effect of relative humidity is small for the Celesco unit and the TSI unit when the precipitator is turned off. When the precipitator was on, the corona action of the test electrode was affected by humidity. This affect was significant above a relative humidity of 35%. Since the corona electrode is present over the test crystal alone, the reference crystal does not effectively compensate for the humidity changes. The level of error at 40% R.H. is shown for the unit in Figure 17. Coating of the electrodes with a layer of silicone grease ( $\sim 5 \text{ } \mu\text{m}$ ) appeared to reduce the effect.

Another source of error is the pick-up of moisture by the hygroscopic deposit. If the particulates and the aerosol are originating at the same environment, this effect would have taken place before the deposition of particles and the deposit would represent the true mass of the particulates in the aerosol.

The collection characteristics of the two units and the results are summarized in Tables 5 to 8. The mass sensitivity begins to decrease above a diameter of  $2 \text{ } \mu\text{m}$  and approaches zero at approximately  $20 \text{ } \mu\text{m}$ . The use of coating material appeared to increase the sensing ability for particles in the  $2\text{-}20 \text{ } \mu\text{m}$  range. Particles significantly greater than  $2 \text{ } \mu\text{m}$ , when present in deposits of polydisperse aerosols, may be effectively sensed.



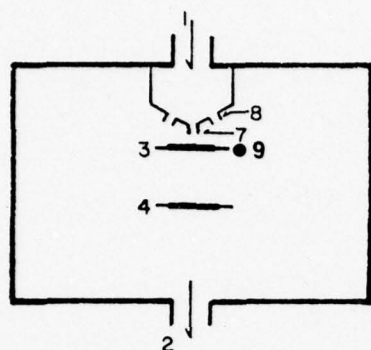


Figure 14: Celesco Schematic Diagram

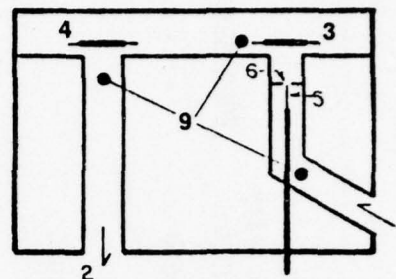


Figure 15: TSI Schematic Diagram

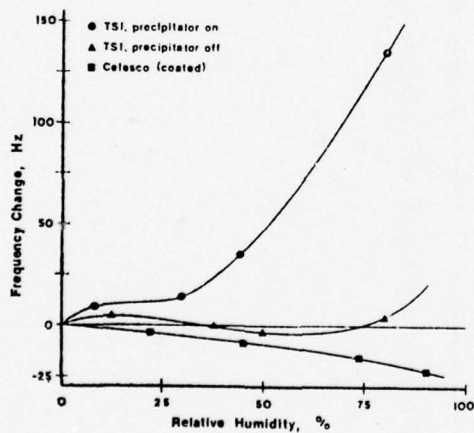


Figure 16: Instrument Responses to Inlet Air Stream Relative Humidity Changes in the Absence of Aerosols

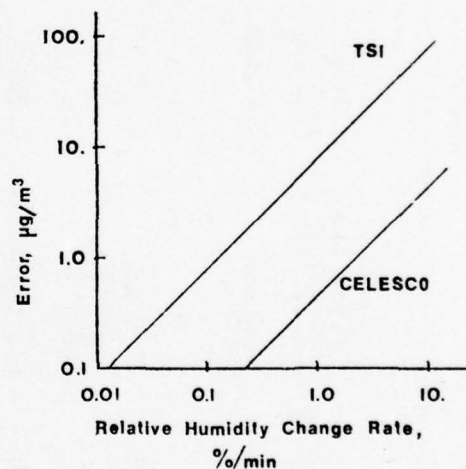


Figure 17: Errors Resulting from Inlet Air Relative Humidity Changes in the Absence of Aerosols ( $e_{hc}$ )



Table 4

## COMMERCIAL QCM MASS MONITORS

Manufacturer	Model No	Deposition Principle	Size Range $\mu\text{m}$	Concentration Range $\mu\text{gm}/\text{m}^3$	Flow Rate lpm	Mass Sensitivity $\text{Hz}/\mu\text{gm}$	Resolution	Operating Time Before Cleaning Crystal
IBC-Berkeley (IBC-Celesco) Irvine, Ca.	PM-40	Impactor	0.3-100*	0-200 0-2000	0.6	$\sim 3000$		few minutes (100 $\mu\text{gm}/\text{m}^3$ )
IBC-Berkeley (IBC-Celesco) Irvine, Ca.	C-100	10-Stage Cascade Impactor	0.3-100*	0-60,000		$\sim 3000$		
Thermo-Systems Inc. St. Paul, Minn.	3205A	Electrostatic Precipitator	0.01-20	1-20,000	1.0	180	+ 1 Hz (+ 0.006 $\mu\text{gm}$ )	1 hr. (100 $\mu\text{gm}/\text{m}^3$ )
	3205B	Electrostatic Precipitator	0.01-20	1-200,000	1.0	180	+ 1 Hz (0.006 $\mu\text{gm}$ )	--

\* Actual upper size value is limited by inlet efficiency to about 30  $\mu\text{m}$ .

Table 5

## TSI MASS MONITOR, POLYDISPERSE AEROSOL RESPONSE SUMMARY

Material	*Particle Size, $\mu\text{m}$	Deposit Diameter, mm	Crystal Sensitivity, Hz/ $\mu\text{g}$	Linear Response Limit			Saturation	
				kHz	$\mu\text{g}$	$\mu\text{g}/\text{mm}^2$	kHz	$\mu\text{g}/\text{mm}^2$
Ariz. road dust	0.3-35	3.8	255	$\cong 0.5$	$\cong 2.5$	$\cong 0.2$	0.8	0.7
Methylene blue-uranine, 9:1, 22% RH, 1.5 mg/m <sup>3</sup>	0.2	5.3	190	0.7	3.5	0.16	2.0	0.5
Ariz. road dust 27% RH	0.3-3	4.3	230	0.7	2.7	0.21	> 3.0	> 1.
(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub> , 27% RH	0.2	5.0	205	0.9	4.5	0.23	1.1	0.3
Laboratory air, $\cong 50\%$ RH	—	5.	205	1.0	5.0	0.25	3.5	0.4
NaCl, 27% RH 1.5 mg/m <sup>3</sup>	0.2	4.3	230	1.0	4.5	0.30	3.5	0.9
Uranine, 45% RH	0.5	5.3	190	1.5	7.4	0.35	—	—
CaSO <sub>4</sub> $\cdot$ $\frac{1}{2}$ H <sub>2</sub> O, $\cong 50\%$ RH	1-3	5.	205	1.5	7.2	0.5	2.5	1.3
Outside air, high visibility, 60% RH	—	5.	205	1.5	7.2	0.5	> 4.	> 1.5
Glycerol	0.5	6.4	155	3.5	23.	0.70	9.5	3.5
Dimethyl polysiloxane, viscosity = 10 <sup>5</sup> cp	0.5	6.4	155	4.1	27.	0.83	9.5	4.
Methylene blue-uranine, 9:1, 22% RH, 0.2 mg/m <sup>3</sup>	0.2	5.3	190	4.0	21.	0.97	20.	> > 3.
NaCl, 27% RH, 0.15 mg/m <sup>3</sup>	0.2	4.3	230	4.0	17.	1.5	4.3	1.6
Dimethyl Polysiloxane, viscosity = 10 <sup>5</sup> cp	0.2	6.4	155	9.0	60.	1.8	9.5	2.0
Uranine, 75% RH	0.5	5.3	190	12.0	63.	3.0	> 15.	> 4.2

\*Ranges estimated optically; 0.2, 0.5  $\mu\text{m}$  aerosols were Collison generated, see text.

Table 6

## TSI MASS MONITOR, MONODISPERSE AEROSOL RESPONSE SUMMARY

Material	Particle Diameter, $\mu\text{m}$	Deposit Diameter, mm	Crystal Sensitivity, Hz/ $\mu\text{g}$	Linear Response Limit				Saturation	
				kHz	$\mu\text{g}$	$\mu\text{g}/\text{mm}^2$	Mono-layers	kHz	$\mu\text{g}/\text{mm}^2$
Polystyrene	0.08	5.0	205	1.6	8.8	0.40	8.0	4.0	1.2
Polystyrene	0.14	4.3	230	1.9	7.5	0.57	7.7	5.4	2.5
Polystyrene	0.41	4.3†	230	3.7	1.45	1.15	4.0	4.2	1.3
Polystyrene	0.65	4.5	220	3.8	15.3	1.10	3.0	4.4	1.2
Polystyrene	1.24	4.0	240	6.0	25.	2.0	2.6	17.	5.
Polyvinyl toluene	1.89	3.4	265	No data					
Styrene divinyl benzene copolymer	7.8*	2.5	285	Not detected efficiently					
Paper mulberry pollen	12.	1.5	300	Not detected efficiently					
Ragweed pollen	20.	1.5	300	Not detected efficiently					
Glass beads	34.	Not collected							

\*Approximately normally distributed with mass mean diameter of 7.8  $\mu$  and standard deviation of 1  $\mu$ .

†Not measured but determined from plot of particle diameter vs. deposit diameter for other sizes.

Table 7

## CELESCO QUARTZ CRYSTAL MICROBALANCE AEROSOL RESPONSE SUMMARY

	Particle	Linear Response Limit				Saturation	
	size, $\mu\text{m}$	Hz	$\mu\text{g}$	$\frac{\mu\text{g}}{\text{mm}^2}$	mono-layers	Hz	$\frac{\mu\text{g}}{\text{mm}^2}$
Monodisperse Materials							
Polystyrene beads	0.41	Not Collected					
Polystyrene beads	0.65	120	0.043	0.35	10.	215	9
Polystyrene beads	0.83	75	0.024	0.22	4.7	275	20
Polystyrene beads	1.24	25	0.009	0.1	1.1	>60	>10
Polyvinyl toluene beads	1.89	30	0.011	0.16	1.3	>300	>20
Polydisperse Materials							
Methylene blue- uranine, 9:1, 27% RH	0.5	75	0.027	0.23	N.A.	>600	>30
Arizona road dust	0.3-3.5	—	—	—	N.A.	150	<20
Uranine, 27% RH	0.5	100	0.036	0.30	N.A.	>400	>70
Uranine, 60% RH (Crystal not coated)	0.5	1900	0.68	5.6	N.A.	>2000	>100

(Crystal not coated)

NOTE: Crystal sensitivity of 2800 Hz/ $\mu\text{g}$  used in calculation of this table. Deposit diameter of 0.4 mm for all materials except 1.24 and 1.89  $\mu$  beads which had 0.35 and 0.3 mm deposits, respectively.

Table 8

## EXPERIMENTAL CRYSTAL MASS SENSITIVITY DETERMINATIONS

Test Aerosol and Technique	Sensitivity, Hz/ $\mu\text{g}$		Sensing Eff., %	
	Runs	Observed† Expected		
Thermo-Systems Unit				
Methylene blue- uranine, 9.2 $\mu\text{m}$ mmd				
Uranine fluorescence	5	273 $\pm$ 16	192	142
Parallel filtration	2	268 $\pm$ 70	192	138
Direct gravimetric	4	198 $\pm$ 22	192	103
NaCl, 0.2 $\mu\text{m}$ mmd				
Parallel filtration	5	265 $\pm$ 10	230	115
1.89 $\mu\text{m}$ polymer beads				
Particle count	1	280 $\pm$ 100	265	105
Arizona Road dust, $\sim$ 2.5 $\mu\text{m}$ mmd				
Direct gravimetric	5	174 $\pm$ 26	192	91
7.8 $\mu\text{m}$ polymer beads				
Particle count	1	45 $\pm$ 15	285	16
12 $\mu\text{m}$ pollen				
Particle count	1	30 $\pm$ 7	300	10
Particle count*	1	36 $\pm$ 20	110	32
20 $\mu\text{m}$ pollen				
Particle count	3	$\sim$ 0	300	$\sim$ 0
Particle count*	1	$\sim$ 0	105	$\sim$ 0
45 $\mu\text{m}$ glass beads	1	Not Collected		
CeleSCO Unit				
Uranine, 0.5 $\mu\text{m}$ mmd				
Fluorescence	4	3075 $\pm$ 1140	2830	108
0.65 $\mu\text{m}$ polymer beads				
Thermo-Systems P.M.M.	3	2950 $\pm$ 1200	2740	107
1.24 $\mu\text{m}$ polymer beads				
Thermo-Systems P.M.M.	3	3500 $\pm$ 1600	2830	123
1.89 $\mu\text{m}$ polymer beads				
Particle count	1	5200 $\pm$ 2000	4800	108
12 $\mu\text{m}$ pollen				
Particle count	1	$\sim$ 0	2830	$\sim$ 0

\*Coated crystal with particle application by settling.

†Limits represent 2 standard deviations of variance for determinations with 3 or more runs or estimated 95% confidence interval for others.

The limit for the linear response was also investigated. For the Celesco unit utilizing impactor sampling, periods were conservatively recommended at a few minutes for a concentration of  $100 \mu\text{g}/\text{m}^3$  and up to an hour for the TSI unit. The difference is due to the size of the deposit area. The mass sensitivity of the crystals was found to be in general agreement with the other results in the literature.

IBC Berkley (formerly IBC-Celesco) also manufactures another instrument which may be used for size analysis as well as for mass measurement [12]. The aerosol sample at a flow rate of 0.25 lpm flows through a 4 to 10-stage impactor designed after the University of Washington reduced-pressure impactor. The quoted particle size range is 0.05 to  $25 \mu\text{m}$ . Each impactor stage is equipped with a QCM. The crystals may be cleaned and/or changed in only a few minutes. At a loading of  $90,000 \mu\text{g}/\text{m}^3$ , the crystal should be cleaned every 10-15 runs of 20 seconds each. The instrument is equipped with a digital output of the fraction mass on each stage. Limitations to this device are a combination of errors discussed for the QCM, and the performance of the cascade impactor which is subject to the errors common to all impactors. These errors include non-uniformity of deposition, loss in the impactor inlet, and body and particle bounce.

The ten-stage impactor is operated at reduced pressure so that the cut-off diameter may be below the conventional  $0.2 \mu\text{m}$  limit. The final stage may have a pressure of only a quarter of an atmosphere. This will affect stability of relatively volatile particles such as aqueous aerosols. The usefulness of the instrument in these situations may be limited to measuring particles above  $0.3 \mu\text{m}$ . The fraction smaller than  $0.3 \mu\text{m}$  usually captured by filter may be lost in this instrument.

## 2.2 Aerosol Size Analyzer

Instruments for aerosol size measurement based on scattering and attenuation of light have been available for a considerable time. These instruments have been limited to relatively dilute aerosols of particle sizes above  $0.5 \mu\text{m}$ . Electrical aerosol analyzers were available for measurement of submicron aerosols. A review of these instruments was presented by Davies [13] from IITRI.

During the past few years, considerable attention had been focused on submicron particles and their role in the environment. Developments in the electrophotical and electronic industry has produced a new generation of instruments with extended capabilities in size and concentration ranges, and automation.



### 2.2.1 Aerosol Size Analyzer Using Conventional Optics

A light beam incident on a particle is scattered in all directions and may be partially absorbed by the particle. The relative amount of light energy scattered in various directions is determined by wavelength of the light, size of the particle, and the refractive index of the particle.

In the optical aerosol counters, the aerosol is sampled through a sensing zone (a viewing volume) so that aerosol particles flow through one by one across an incident light beam. The light scattered by the particles is detected by a photodetector such as a photomultiplier tube and a signal in form of a photocurrent is generated. This signal is then processed to produce a well-defined pulse whose amplitude is proportional to radiation scattered by the particle into the photodetector. As the particles flow sequentially through the sensing zone, a series of the voltage pulses are generated and are classified into several categories based on the magnitude of the signal. The number of particles of a given size class is then displayed and/or recorded so that a frequency distribution may be computed.

The commercial photometer based on conventional optics is useful for measurement of particles above 0.5  $\mu\text{m}$ . The upper size is usually limited to below 50  $\mu\text{m}$  due to sampling errors associated with large particles.

Cooke and Kerker [14] have presented calculations for the theoretical photometer response based on the instrument geometry and principle. Aerosol analyzers may be divided into two classes depending on their scattering geometry. The first system is based on the collection of light symmetrically scattered around the axis of the incident illumination. The second system is based on collection of light scattered around a central angle by an off axis collector. These two systems are shown schematically in Figure 18.

The photometer response is given by

$$R = \iiint \frac{\lambda^2}{8\pi^2} (i_1 + i_2) F(\lambda) F(\theta, \phi) d\lambda d\theta d\phi \quad (5)$$

where  $\lambda$  is the wavelength of light,  $i_1$  and  $i_2$  are the angular intensity functions,  $f(\lambda)$  is the wavelength distribution of the emissive power light source combined with the spectral sensitivity of the phototube, and  $F(\theta, \phi)$  is a geometrical factor depending on the geometry of the incident light beam and the collection angle. The scattering angle  $\theta$  is measured with respect to the incident ray and  $\phi$  is measured between the incident beam and the illumination axis.

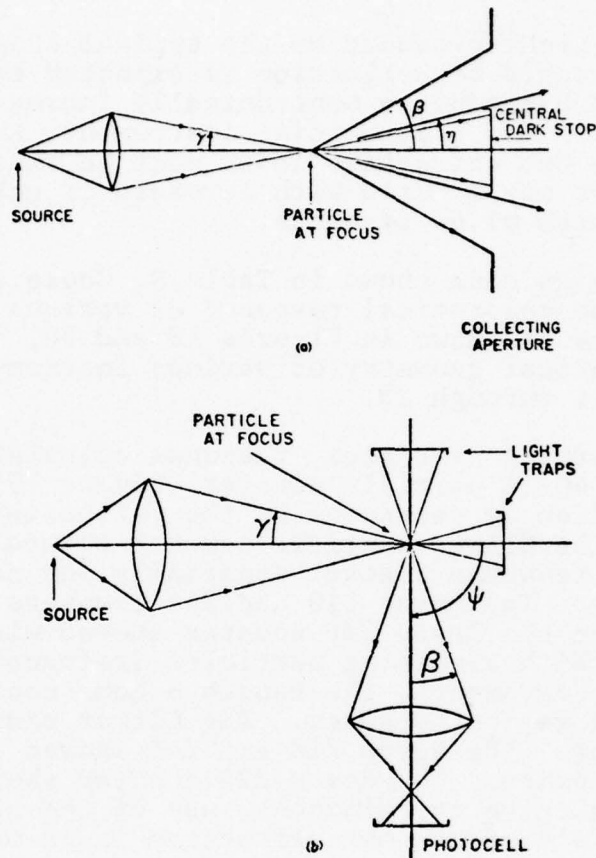


Figure 18: (a) Particle counter geometry for convergent illumination (semiangle  $\gamma$ ), with a collecting aperture (semiangle  $\beta$ ) having a central dark stop (semiangle  $\eta$ ). (b) Particle counter geometry for convergent illumination (semiangle  $\gamma$ ) with a collecting aperture (semiangle  $\beta$ ) inclined at an angle  $\psi$  to the axis of the incident radiation.

The angular intensity function  $i_1$  and  $i_2$  depend upon the particle size parameter  $\alpha = 2\pi r/\lambda$ , and the refractive index  $m$  (which may have an imaginary part for absorbing particles). These intensity functions are oscillatory functions at a given angle and a single measurement may represent multiple values of the size parameter.

Use of white light produced by the typical tungsten filament and the wide angle of collection is expected to damp out the oscillations and produce a monotonically increasing response with particle size. For a commercial instrument, the dependence of the response on the refractive index must be minimized so that the instrument may be used with aerosols of unknown refractive indices and with mixed aerosols.

Using the design data shown in Table 9, Cooke and Kerker [14] have calculated the theoretical response of various light scattering counters as shown in Figures 19 and 20. Schematic drawings of the optical geometry of various instruments is shown in Figures 21 through 25.

With non-absorbing particles, response calculations for the Bausch & Lomb 40-1A particle counter (Figure 19) show that the response is strongly dependent on the refractive index and is multivalued. The Climet particle counter showed a much narrower range of response scatter especially for particles above  $1\text{ }\mu\text{m}$  in size. The Royco 218 and 245 counters showed multivaluedness and the Royco 220 counter showed wide scatter in the response. With absorbing particles (refractive index with an imaginary component), the Bausch & Lomb counter showed multivaluedness as well as scatter. The Climet counter also showed wide scatter. The Royco 218 and 245 showed less wide scatter of the response. The Royco 220 counter showed wide scatter. Based on these calculations, use of the commercial counters on materials of unknown refractive index may lead to significant errors.

At the University of Minnesota, the response characteristic of the commercial counters were experimentally investigated using monodisperse aerosols. Liu, Berglund, and Agarwal [15] and Willeke and Liu [16] have discussed the findings of these investigations. Bausch & Lomb 40-1A, Royco 220 and Royco 245 were investigated. The Model 220 is a  $90^\circ$  scattering instrument while the 245 and 40-1A are forward scattering instruments as shown in Figures 26 through 28.

The Bausch & Lomb 40-1A produces a monotonically increasing response for di-octyl-phthalate (DOP) aerosol which is commonly used in the calibration of particle counters. The Royco 245 showed a dip at around  $1\text{ }\mu\text{m}$  indicating that the size measurement below  $1\text{ }\mu\text{m}$  may not be reliable. The Royco 220 showed a significant change in response with refractive index. The effect of

Table 9

## OPTICAL PARAMETERS OF COMMERCIAL SIZE ANALYZERS

Manufacturer	Model No.	Angle $\gamma$	Angle $\eta$	Angle $\beta$	Sample Volume cm <sup>3</sup>	Concentration limit (particles/cm <sup>3</sup> )	
						1% coincidence	10% coincidence
Bausch & Lomb	40	13	33	53	$5 \times 10^{-4}$	20	210
Climet	208	15	35	90	$5 \times 10^{-4}$	20	210
Royco (90°)	220	24	--	24	$2.63 \times 10^{-3}$	4	40
Royco	245	5	16	25	$4.0 \times 10^{-3}$	2.5	25
Royco	225	5	16	25	$5 \times 10^{-4}$	20	210
Tech Ecology	208	5	10	20	$2.5 \times 10^{-3}$	4	40



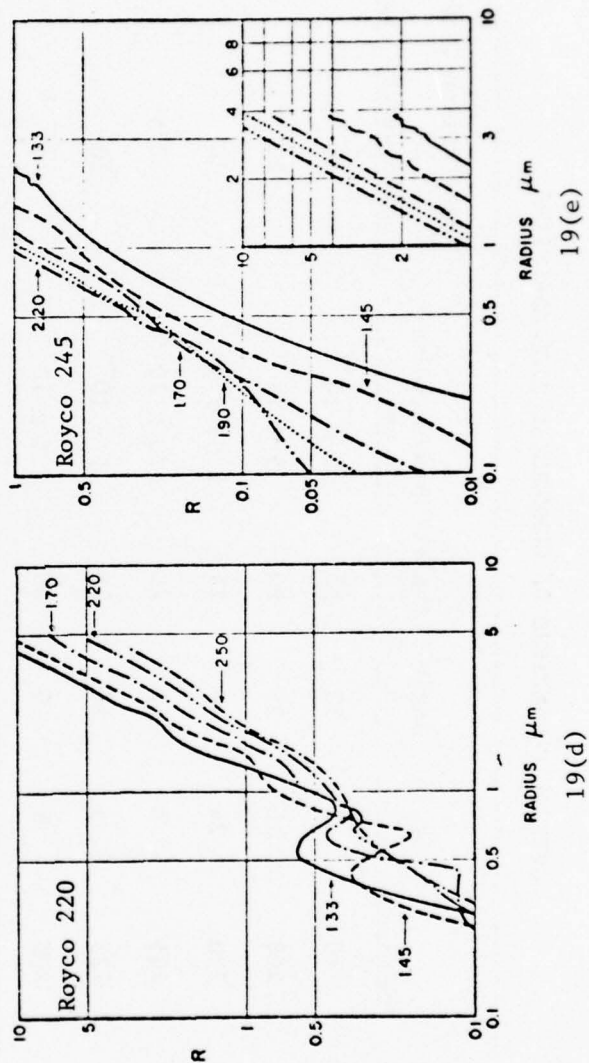
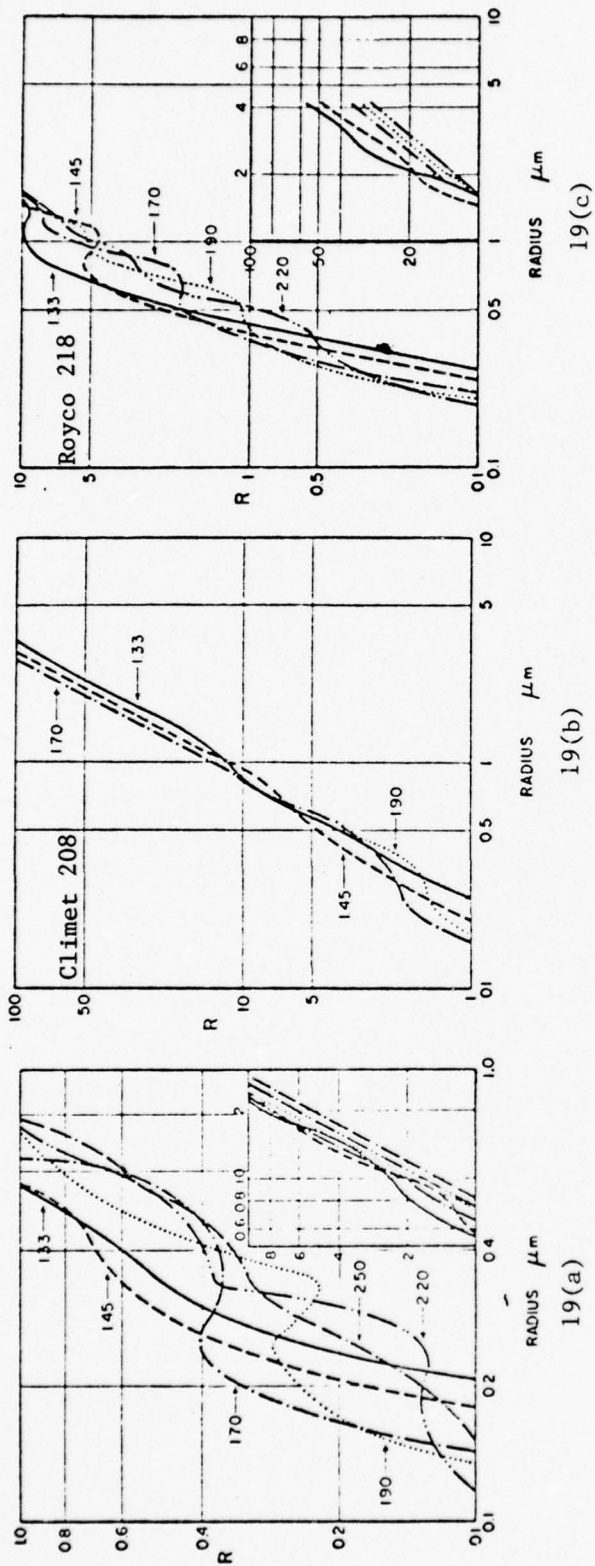


Figure 19: Theoretical Response - Non-Absorbing Particles

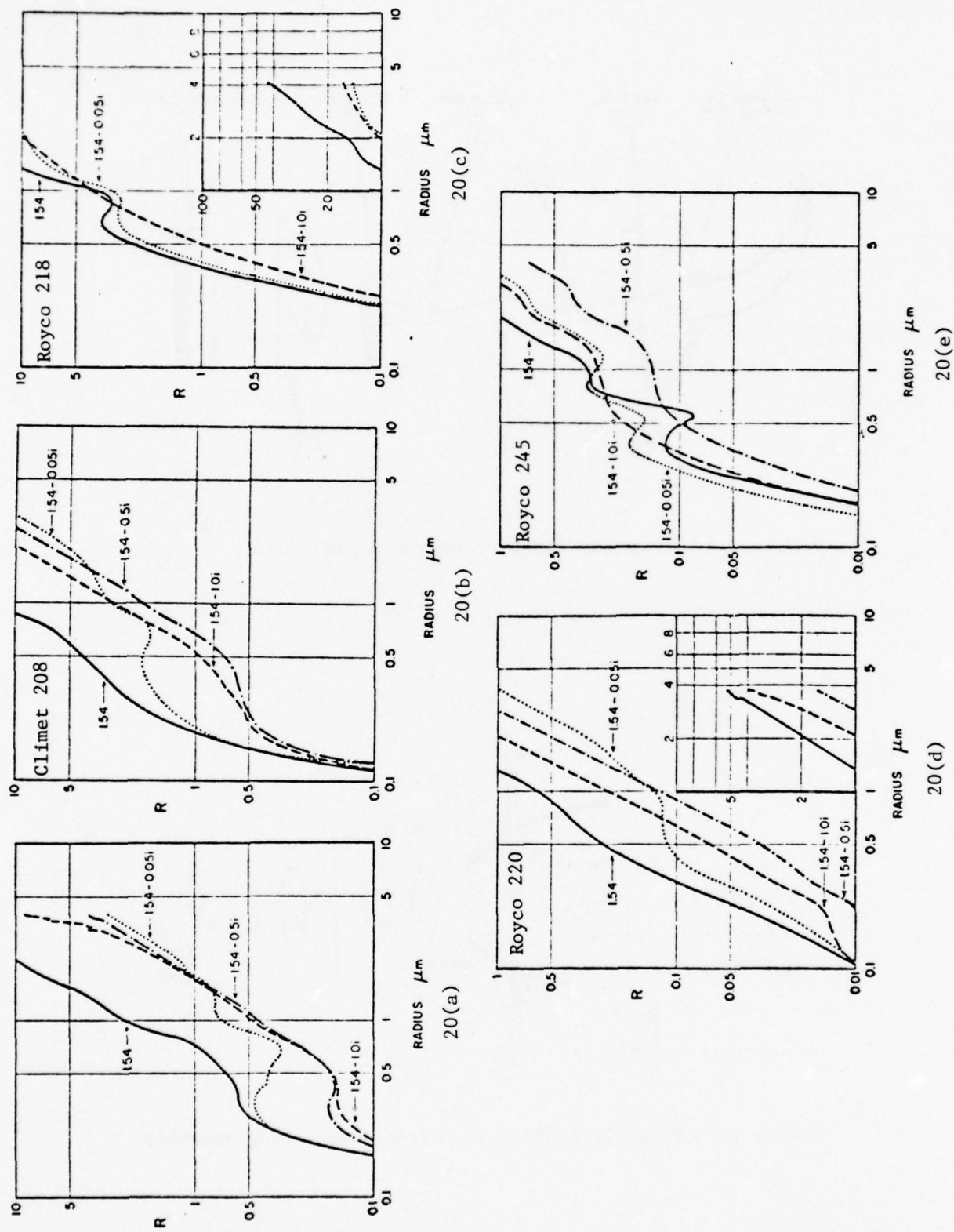


Figure 20: Theoretical Response - Absorbing Particles

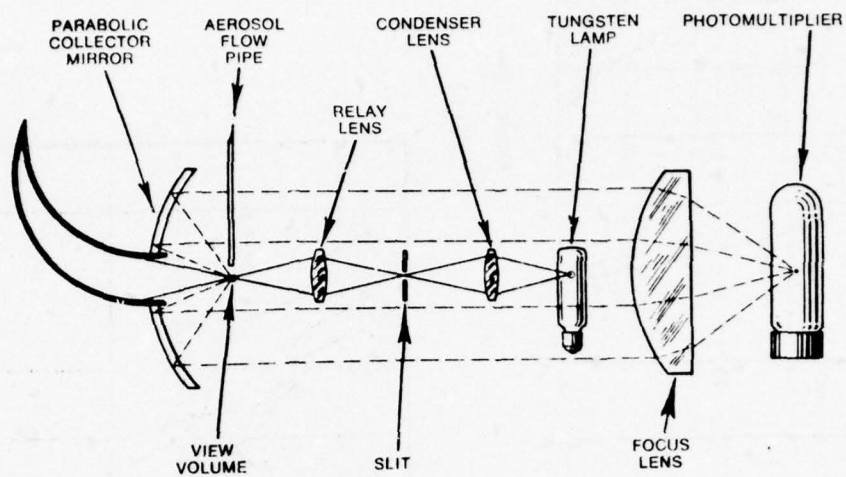


Figure 21: Schematic Diagram; Bausch & Lomb 40-1A.

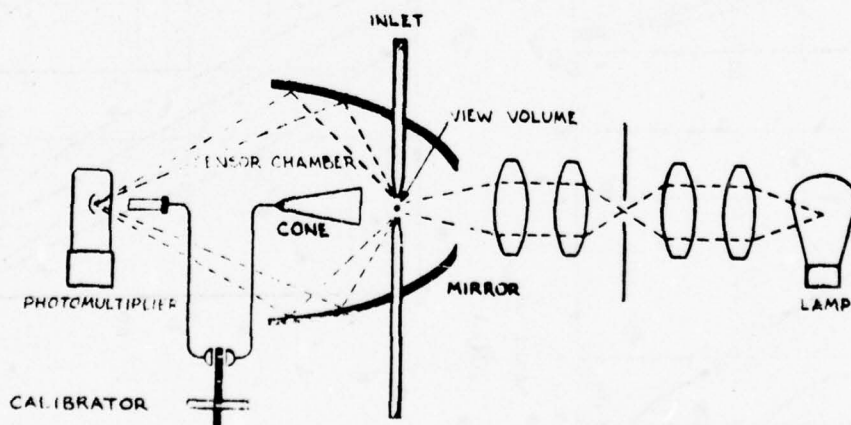


Figure 22: Simplified Diagram, Particle Detecting Assembly.

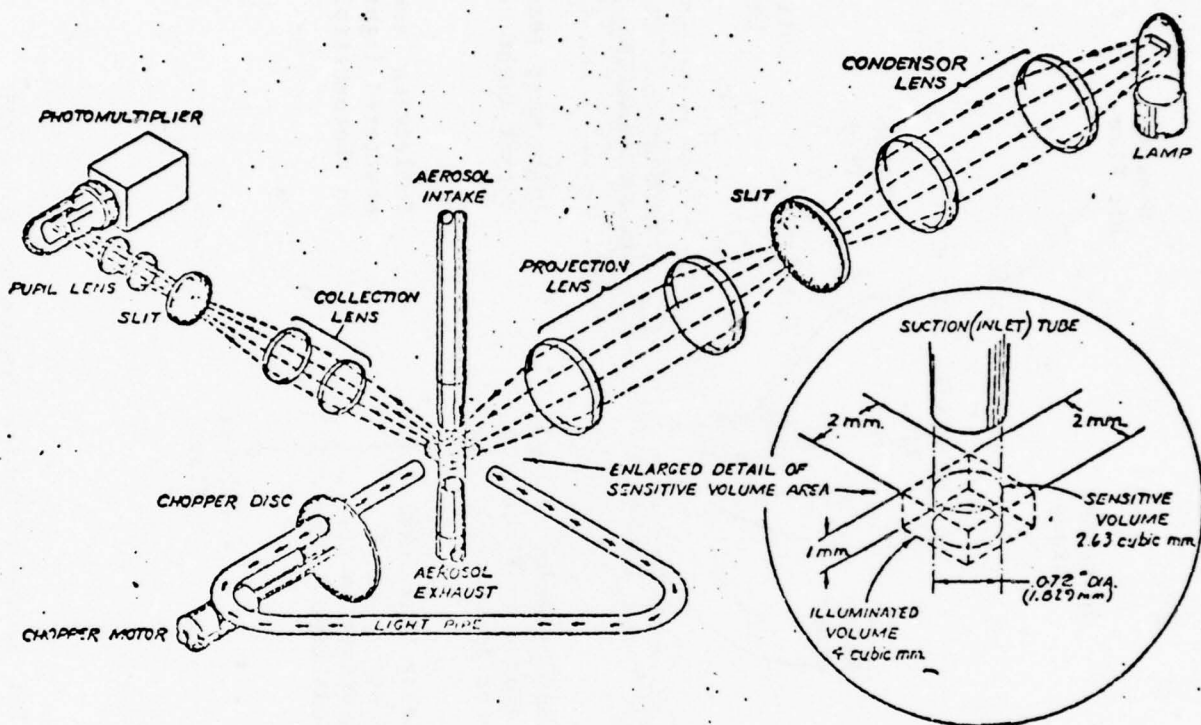


Figure 23: Schematic Diagram of the Royco 220 Optical System from the Royco 220 Operation and Maintenance Manual



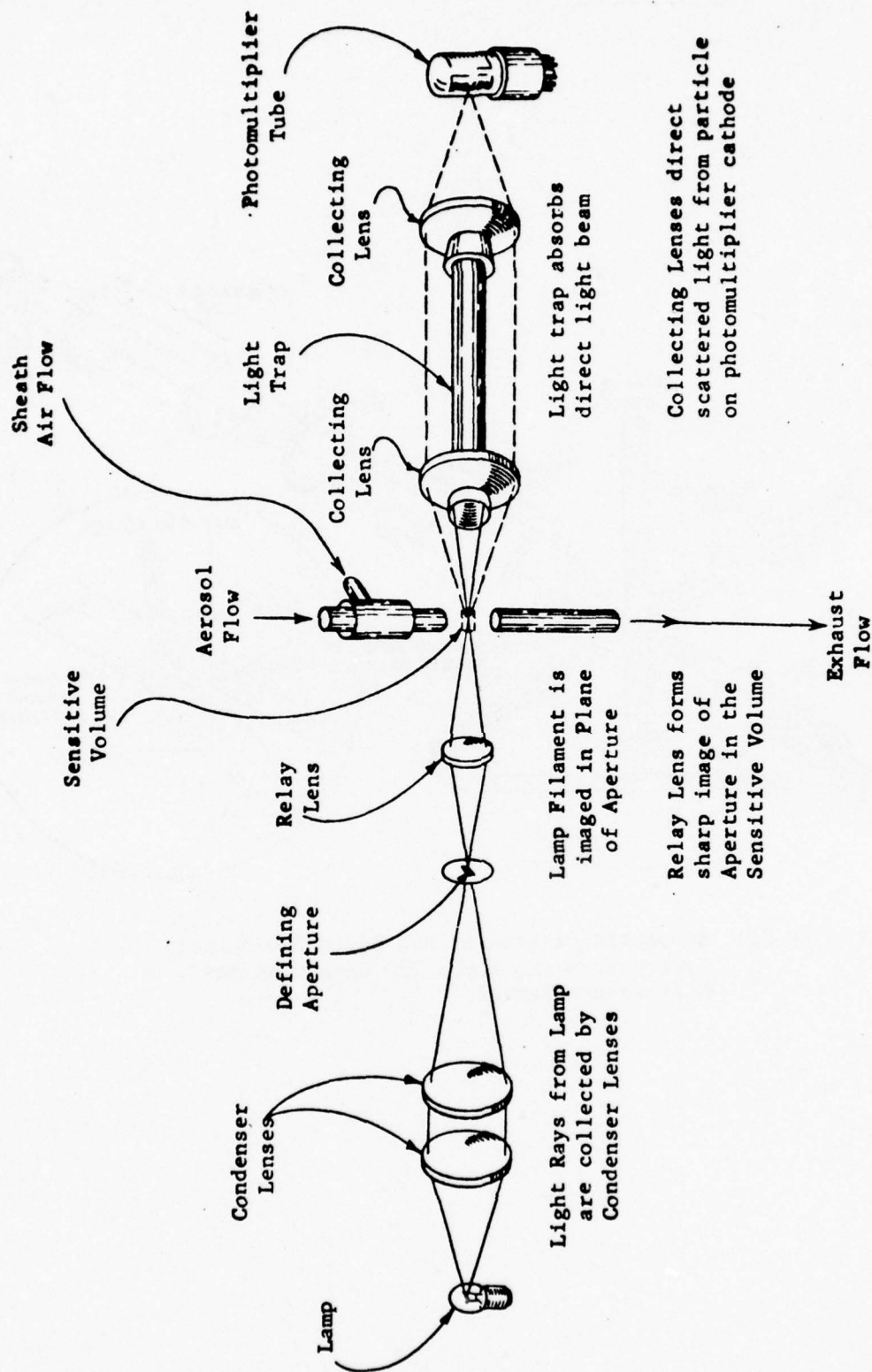


Figure 24: Layout of Optics Components, Royco 225

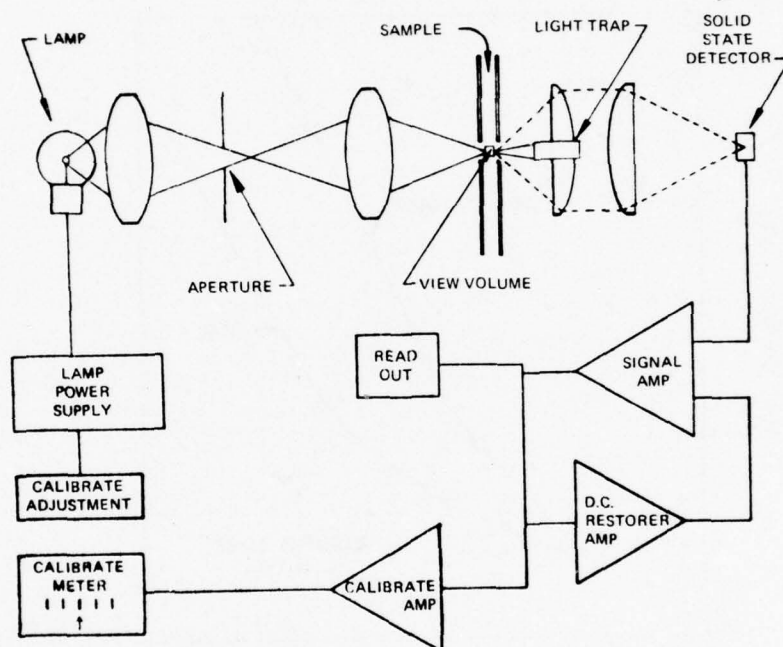


Figure 25: Schematic Drawing: Tech Ecology 208.

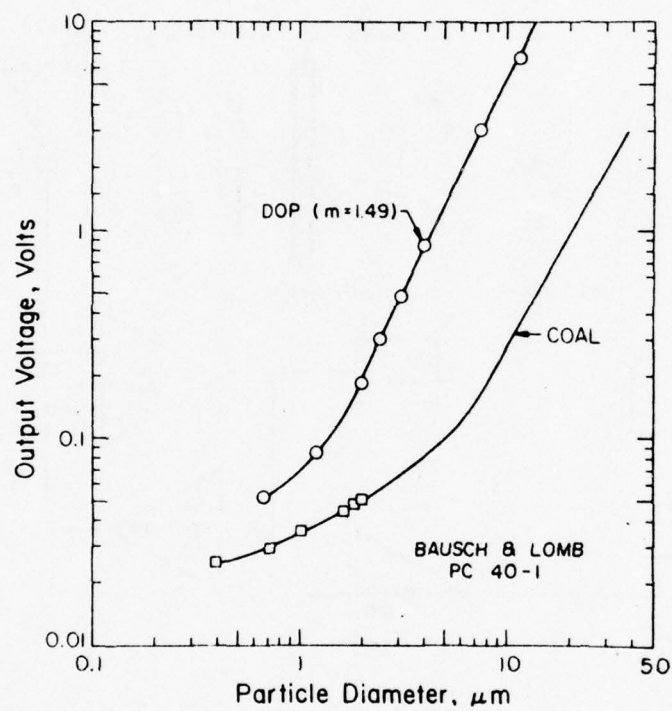


Figure 26: Experimental calibration curve for the Bausch & Lomb 40-1A particle counter.

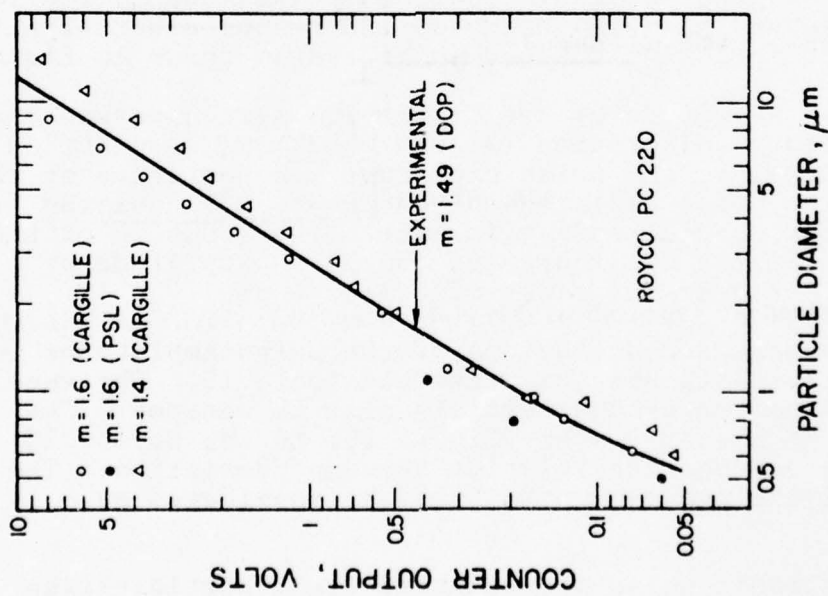


Figure 27: Experimental calibration curve for the Royco 220 particle counter.

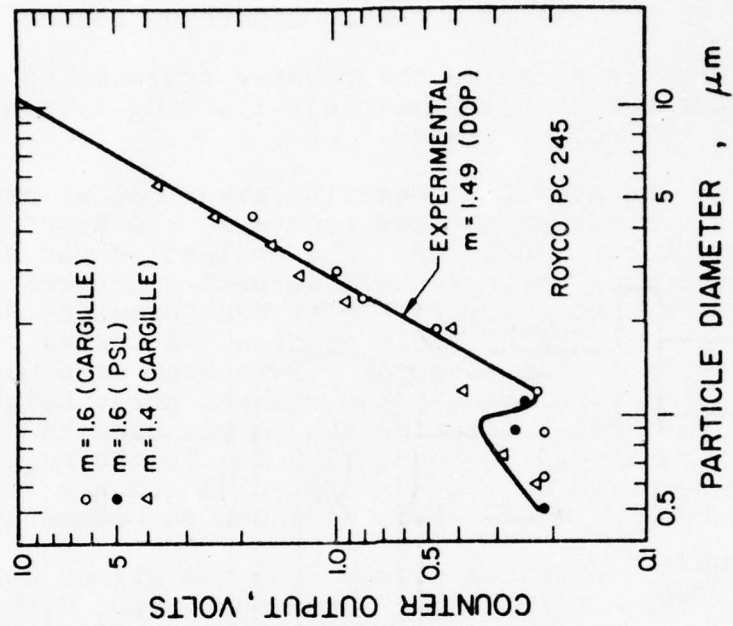


Figure 28: Experimental calibration curve for the Royco 245 particle counter.



the refractive index may be taken into consideration by a "shift factor" for the response at refractive indices other than the DOP aerosol used for calibration. As seen in Figure 28, the shift factor increases from three for small particles to about five for large particles when the aerosol material was changed from DOP ( $m = 1.49$ ) to a highly absorbing carbon aerosol.

Effect of particle shape on the counter response is expected to be less for an axisymmetric collecting system rather than the off axis collection system.

Eaton, Clark, and Avol [17] describe experimental evaluation of two light scattering optical counters, the Royco Model 220 and the Climet Model 208. The evaluation was a part of an EPA sponsored program to install aerosol concentration and size measurement instrumentation with EPA Community Health Air Monitoring Program (CHAMP) field station. A series of monodisperse aerosols in the range of 0.3 to 6  $\mu\text{m}$  were used. The response was processed using a 256 channel pulse height analyzer. The Climet 294-1 dilution system was used to dilute the aerosols at a ratio of 100:1 and 1000:1. The Royco counter was evaluated as received and with modified sheath air inlet system described by Liu, et al. [15] as shown in Figure 29.

The shape and width of the pulses from the Climet 208 were uniform over the entire range. Spurious pulses were observed with the basic unit with and without the dilution system. These pulses were thought to result from recirculation of particles in the viewing volume. The Royco 220 produced pulses which were uniform in shape but had a width which depended on the pulse height. Spurious pulses due to recirculation were observed also in the Royco 220 response but significantly reduced following the Liu inlet system shown in Figure 29.

The pulse amplitude of the Climet 208 agreed reasonably well with factory calibration (within + 22%). The dilution system did not alter the pulse amplitude for particles of the same size. The pulse amplitude of the Royco 220 deviated from calibration for particles 2  $\mu\text{m}$  in size. The addition of the sheath inlet (Figure 29) increased the pulse amplitude by 20 to 43 percent over the range of 0.3 to 6  $\mu\text{m}$ .

Relative standard deviations of the pulse amplitudes for monodisperse aerosols are summarized in Table 10. The resolution becomes poorer as the particle size is reduced. The addition of the sheath system (Figure 29) to the Royco 220 significantly reduces the relative standard deviation. The resolution of the two instruments may be considered as comparable.

The electronic noise was measured using particle-free air. The noise levels for both instruments exceeds the pulse

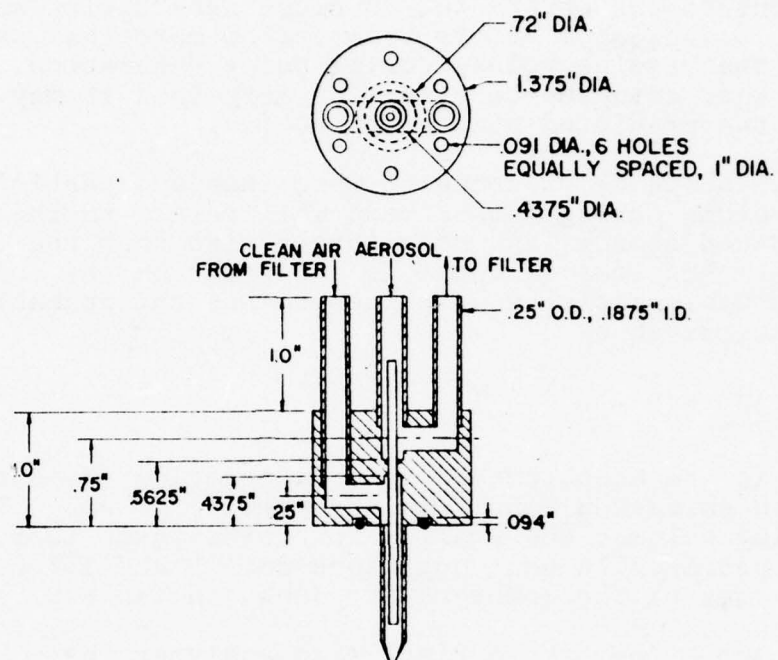


Figure 29: Sheath-air inlet for the Royco 220 particle counter.

height corresponding to 0.3  $\mu\text{m}$  particles. The noise contribution by the Climet counter, the Royco counter, and the Royco with the sheath air inlet are 0.706, 0.035, and 0.624 particles/ $\text{cm}^3$  ( $2 \times 10^4$ ,  $10^3$ ,  $1.7 \times 10^4$  particles/ $\text{ft}^3$ ) respectively at flow rates of 118, 47.2, and 0.608  $\text{cm}^3/\text{sec}$ , respectively.

The performance of the optical counter is also affected significantly by cross sensitivity of the channels used in the pulse-processing circuits. Janicke [18] and Willeki and Liu [19] have discussed the errors due to cross sensitivity and coincidence. Coincidence errors result from more than one particle being in the viewing volume during pulse generation. Janicke suggests that coincidence errors of more than 1% may affect the shape of the predicted size distribution.

Coincidence or presence of more than one particle in the viewing volume during measurement will result in the particles being counted as one; and of a larger size than the individual particles. The loss of count is dependent on the concentration of the aerosol particles which determines the probability of coincidence given by

$$N_o = N \cdot e^{-N \cdot v} \quad (6)$$

where  $N_o$  is the measured number concentration,  $N$  is the true number concentration, and  $v$  is the viewing volume. The smaller the viewing volume, the smaller the coincidence loss. Calculations of permissible concentrations for 1% and 10% coincidence loss for some of the counters are shown in Table 9.

In conclusion, the optical size analyzers have been studied and developed extensively. The user of these instruments should check the documented performance against the intended use. In particular, the refractive index and concentration conditions must be considered in the choice of the instrument and its operation.

Most of the instruments are well automated and various options of output and display formats are available. The flow rates range from 0.1 to several  $\text{ft}^3/\text{min}$ .

### 2.2.2 Forward Scattering Ratio-Type Laser Light Scattering Size Analysis

Kreickenbaum and Shofner [20] described an aerosol analyzer based on laser optics. A schematic diagram of the unit manufactured by ESC, Inc. is presented in Figure 30.

A 0.94  $\mu\text{m}$  laser is used as the light source. The intensity of light scattered by a single particle in the viewing volume is simultaneously sensed at two angles ( $14^\circ$  and  $7^\circ$ ) in the forward scattering lobe. The viewing volume is  $2 \times 10^{-7} \text{ cm}^3$ .

Table 10

RELATIVE STANDARD DEVIATION OF PULSE AMPLITUDES FOR MONODISPERSE PARTICLES  
FOR THE CLIMET 208 AND ROYCO 220 COUNTERS

Monodisperse Particle Size ( $\mu\text{m}$ )	Relative Standard Deviation of Pulse Amplitude, %			
	Climet 208	Climet with Climet 294 Dilution System	Royco 220	Royco with Univ. Minn. Sheath-Air Inlet
0.5	23.6	23.4	27.0	15.9
0.6	22.2	--	25.3	13.4
0.714	18.8	17.3	24.7	9.90
1.01	14.7	13.8	22.8	9.21
2.02	7.38	7.69	20.7	6.73
3.23	5.40	--	--	5.66
5.49	5.50	--	--	4.26



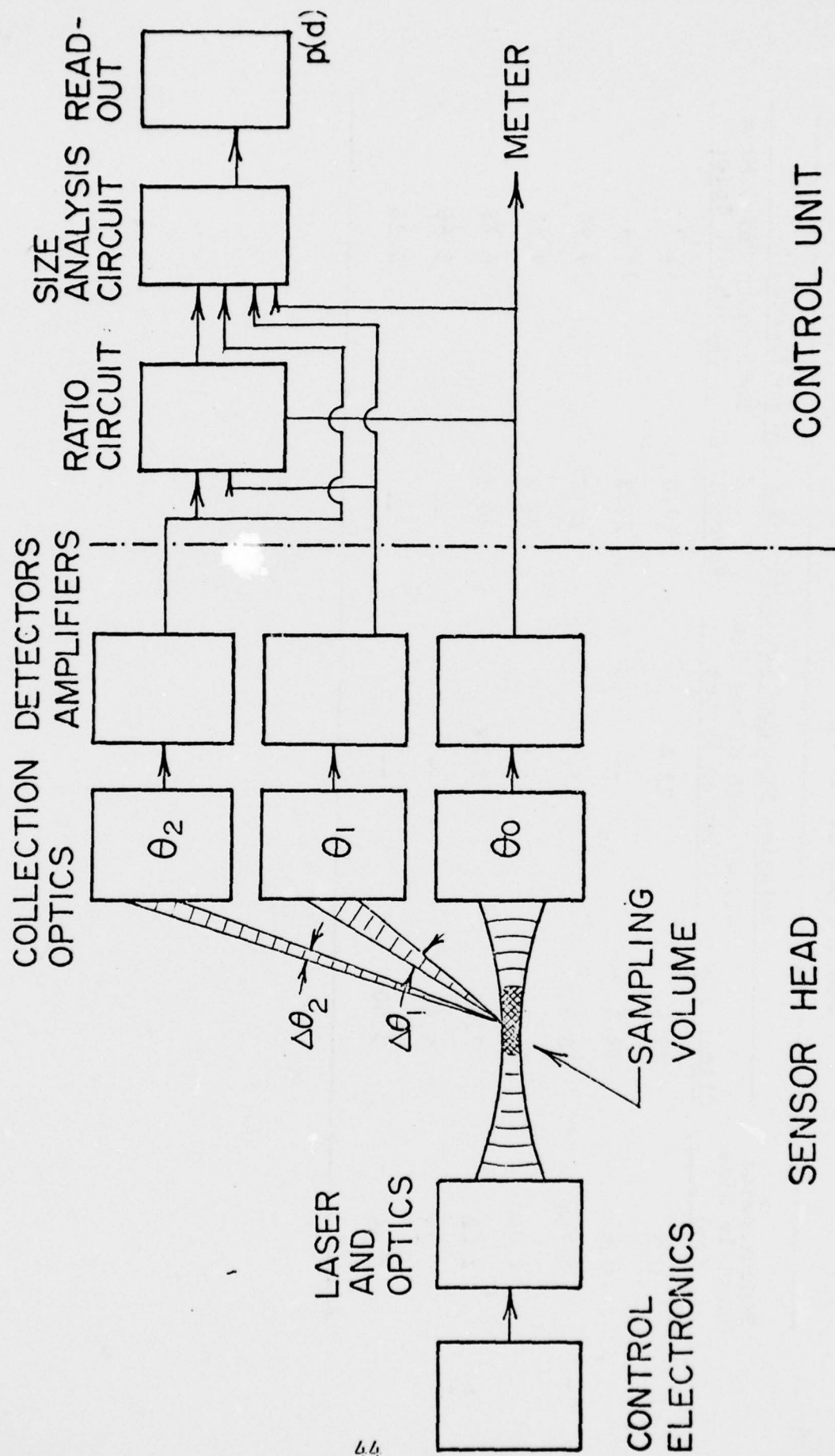


Figure 30: Schematic Diagram: PILS-IV

The manufacturer quotes  $10^6$  particles/cm<sup>3</sup> for  $d < 0.2 \mu\text{m}$  as the upper limit of concentration. This concentration level is typical of a fairly concentrated aerosol and is better than claimed by any other scattering instrument. Using equation 6 for coincidence loss, we find that

$$\frac{N_o}{N} = e^{-10^6 \cdot 2 \cdot 10^{-7}} = e^{-0.2}$$

or

$$\frac{N_o}{N} = \frac{1}{1.223} = 0.82$$

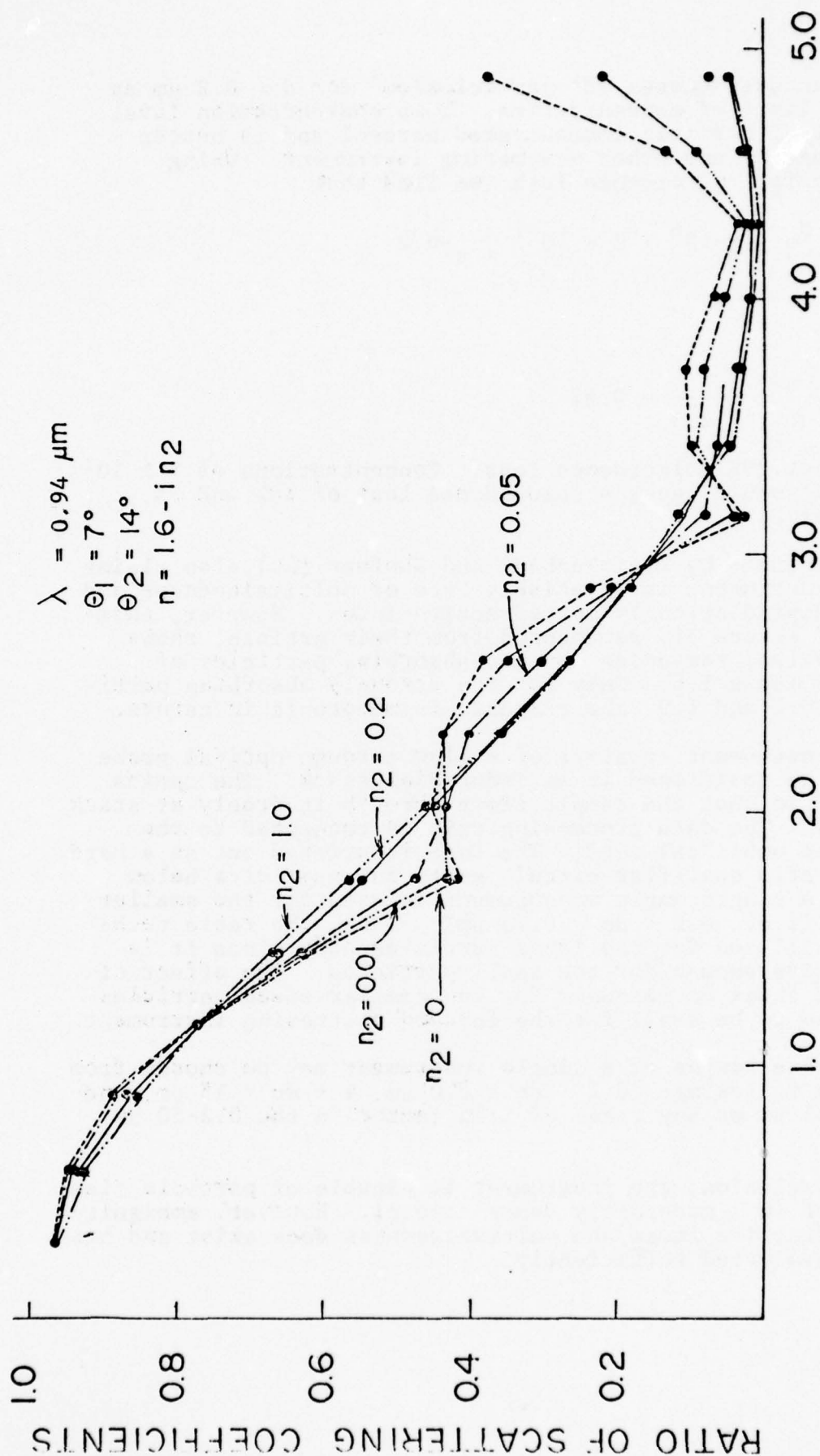
i.e., almost 20% coincidence loss. Concentrations of  $5 \times 10^5$  and  $5 \times 10^4$  would cause a coincidence loss of 10% and 1%, respectively.

The article by Kreickenbaum and Shofner [20] also claims that the instrument is relatively free of multivaluedness and does not depend strongly on refractive index. However, examination of Figure 31, reproduced from their article, shows multiple valued responses for non-absorbing particles of refractive index 1.6. Only for the strongly absorbing particles  $n_2 = 0.2$  and 1.0, the response is monotonic in nature.

The instrument consists of a flow through optical probe which may be positioned in an industrial stack. The optics is mounted so that the sample flows through it freely at stack conditions. The data processing unit is connected to the probe by an umbilical cord. The data is printed out as a hard copy. A ratio qualifier circuit sorts out particles below  $0.75 \mu\text{m}$ . A single angle measurement is used for the smaller particles (i.e.,  $0.2 < dp < 0.75 \mu\text{m}$ ). Thus, the ratio technique is utilized for the large particles only since it is not sensitive enough for the small particles. The effect of refractive index on response for the smaller sized particles is expected to be small for the forward scattering instrument.

The size ranges of a single instrument may be chosen from one of the following:  $0.2 < dp < 3.0 \mu\text{m}$ ,  $1 < dp < 15 \mu\text{m}$ , and  $3 < dp < 45 \mu\text{m}$  or any range of 1:15 factor in the  $0.2$ - $50 \mu\text{m}$  range.

In conclusion, the instrument is capable of particle size measurement in a moderately dense aerosol. However, ambiguity on the refractive index and multivaluedness does exist and has not been evaluated sufficiently.



PARTICLE DIAMETER, micrometer

Figure 31: Theoretical Response: PILS-IV

### 2.3.3 Active Scattering Aerosol Spectrometer

The active open cavity of a gas laser provides an intense particle illumination source for light scattering measurements in the submicron range. Work describing single particle extinction within the laser cavity was first described by Schleusener [21] and later by Schuster and Knollenberg [22]. Knollenberg and Luehr [23] have described the commercial instrument providing a noise analysis and theoretical scattering response.

An Active Scattering Aerosol Spectrometer is one that uses the active open cavity of a laser as the source of particle illumination. The interferometric aspects of the oscillating radiation illuminating the particles produces both forward and backward scattered radiation at all collecting angles. This, coupled with the fact that the collecting optics solid angle can be considerably greater than two steradians, results in a system with great particle sensitivity and reduced sensitivity of refractive index.

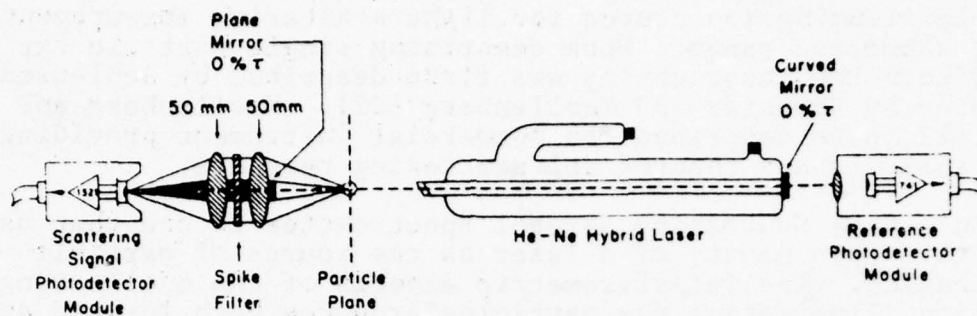
The particles are illuminated with a source of radiation many times greater than that possible with the hottest incandescent source known. This results in a system fully capable of sizing particles several hundred angstroms diameter using photomultiplier detectors or 0.1 microns using solid state silicon detectors. The fundamental limit of size detection is governed by particle evaporation rather than the noise component of the scattering from the gas molecules.

By using a beam splitter and double pulse height analysis, a system has been constructed which determines where a particle passes through a beam in the sampling volume, and reduces background light. The entire sampling volume has unrestricted flow precluding problems with evaporation of wet aerosols and the effects of plumbing walls.

The optical geometry is shown in Figure 32. At the sample volume location, shown in Figure 33, the laser beam is approximately 500  $\mu\text{m}$  in diameter. A centerline brightness in excess of 1000  $\text{w}/\text{cm}^2$  is obtained. Theoretical response of the instrument is shown in Figures 34 and 35. Due to the large collection angle and the integrated forward and back scatter, the response is relatively free of dependence on the refractive index.

The Active Scattering Aerosol Spectrometer (ASAS) is manufactured by PMS, Inc., Boulder, Colorado. The ASAS instrument is designed to size particles into fifteen linear size intervals, with a range change switch to provide up to four independent size ranges within a single system. Other features include trialkali photomultiplier detectors for ultimate sensitivity and "free flow aspiration" for absolute "in situ" sampling. It is packaged in two separate enclosures housing the probe with its optics and pre-amps and an electronics console.





Note: Mirrors are reversed for large particle size ranges.

Figure 32: Schematic of active scattering technique system using an open cavity laser.

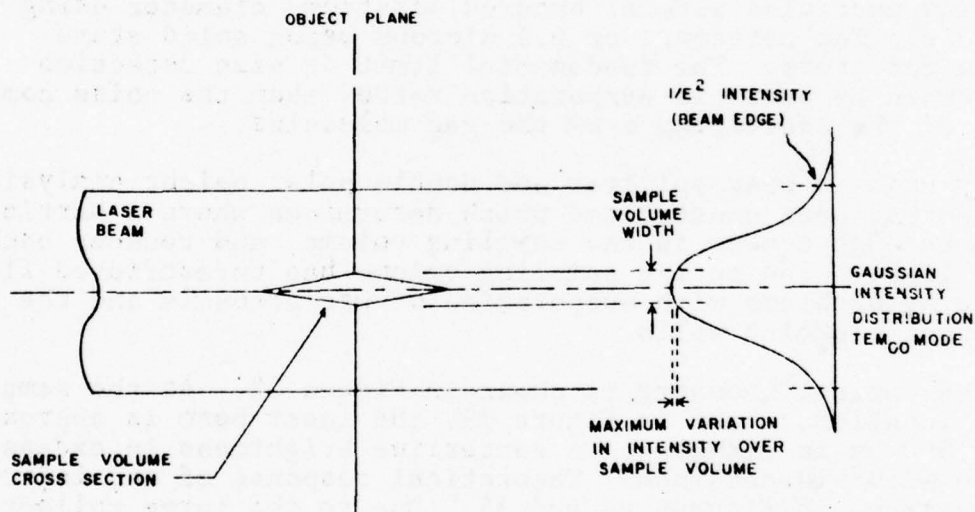


Figure 33: Relative size and position of sample volume cross section.

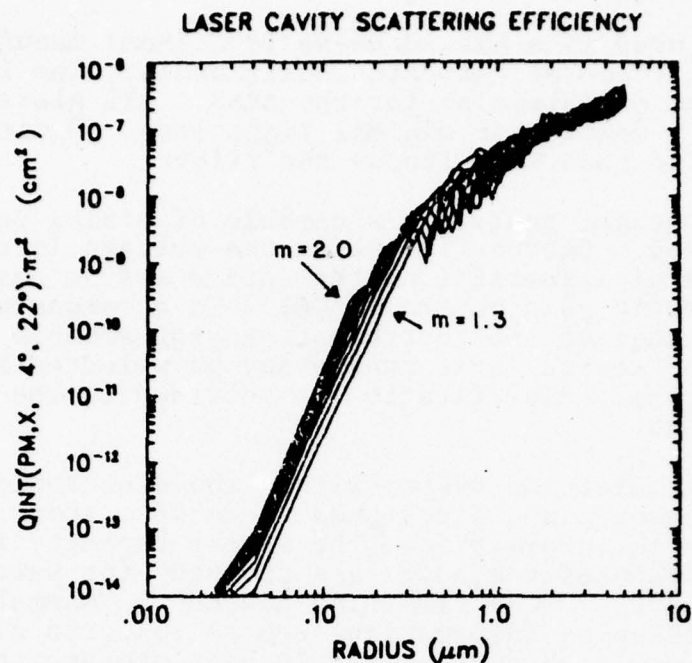


Figure 34: Theoretical instrument response for the  $4^\circ$ - $22^\circ$  collecting geometry for  $m = 1.3$ ,  $m = 1.4$ ,  $m = 1.6$ ,  $m = 1.7$ ,  $m = 1.8$ ,  $m = 1.9$ ,  $m = 2.0$ .

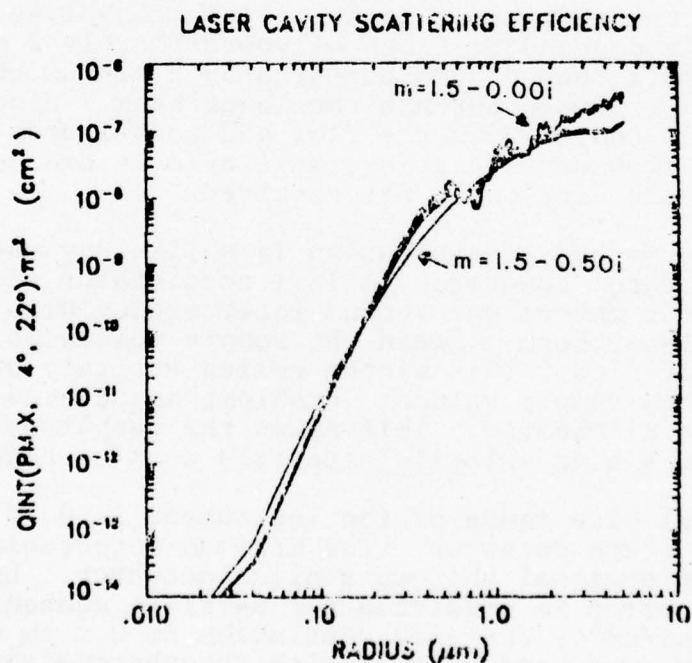


Figure 35: Theoretical instrument response for absorbing particles for  $4^\circ$ - $22^\circ$  for  $m = 1.5$ , and imaginary components of  $i = 0.5$ ,  $0.1$ ,  $0.08$ ,  $0.06$ ,  $0.04$ ,  $0.04$  and  $0.00$ .

The laser used is a hybrid He-Ne (632.8 nm) manufactured by Coherent Radiation of Palo Alto, California. The laser has optimized parameter selection for the ASAS. All glass optical elements are A-R coated for minimal light loss. Optical transmission is better than 90% without the filter.

The pulse height analyzer is capable of sizing pulses as short as 100 nsec. Because its reference voltage is derived from the source of illumination, the entire system has an effective automatic gain control (AGC). To accommodate the large dynamic range of the instrument, a programmable amplifier is used and several size ranges may be selected by a switch. Logarithmic amplification is provided in the tenth micron size range.

The data acquisition system within the electronics console has an active memory and is designed to decode, store, and display particle size information. The memory capacity is sixteen addresses. Fifteen of the addresses are used for particle size distribution storage. The remaining address is normally used for other housekeeping information such as selected size range and elapsed time. Each address has 16 bits storage capacity for 0-9999 counts BCD. A selectable digital display and a graphical CRT display are provided for real-time data monitoring requirements of precise particle counts or distribution functions. An optional printer is available for obtaining a hard copy of the data.

Two types of aspiration are used. The first type used primarily with the reflecting optical system involves a pair of diametrically opposed orifices of approximately 2 mm inside diameter. A small vacuum pump buffered by a plenum chamber draws the aerosol sample through the laser beam. Since the sample volume is centered in the flow and constitutes a few percent of the flow cross section, wall effects are considerably reduced and sheath airflow is not required.

The second method of aspiration is unlike any used in conventional aerosol counters. A 10:1 accelerator produces a flow rate of 6.15 meters per second intersecting with a 3 cm length of the laser beam. Again the sample volume is positioned at the center of flow. This system relies entirely upon optical definition of the sample volume. Problems associated with small bore tubing are eliminated. This makes the technique particularly useful for sizing volatile materials such as natural smog.

The nominal size range of the instrument is 0.15 to 4.5  $\mu\text{m}$  with the solid state detector. The minimum detectable size is 0.08  $\mu\text{m}$  with an optional photomultiplier detector. Less than 1% coincidence error is predicted for particle concentration of  $10^3$  particles/ $\text{cm}^3$ . The size resolution is 0.1  $\mu\text{m}$  with the solid state detector, and 0.05  $\mu\text{m}$  with the photomultiplier tube. Digital and CRT displays and digital and analog output are available on the standard instrument.

The standard solid state detector is better than the photomultiplier detector from noise considerations. The advantages of the photomultiplier exist only at the low signal levels expected from particles less than 0.1  $\mu\text{m}$  in size.

In conclusion, the PMS active scattering instrument has a capability of measuring flowing particles in the 0.1 to 5  $\mu\text{m}$  range without major errors due to refractive index of the particle (sample). Fairly concentrated aerosols, up to  $10^4$  particles/ $\text{cm}^3$  can be analyzed. Since the unit has been on the market for only a couple of years, complete evaluation in actual use is not possible at present. However, it is being used extensively in several aerosol laboratories and general remarks have been very encouraging. It is anticipated that within a couple of years, the instrument will be considered a standard.

#### 2.2.4 Spectral-Particle Counter

The Sartorius-Membrane Filter has introduced an instrument based on scintillation spectral analysis of the particles to obtain size and concentration information.

The instrument has been described by Binke [24]. He compared the spectral analyzer and a  $45^\circ$  light scattering technique and concluded that an order of magnitude higher sensitivity for the spectral analyzer was possible.

In the Sartorius instrument [25], the aerosol particles are brought individually into a heating chamber containing a burner for a hydrogen-air flame (Figure 36). This is a micro double flame of elliptical cross section which provides extremely stable thermal conditions. The atoms or molecules of the particle mass are heated in the flame to their excitation temperature and emit, during their stay in the flame, a flash of light whose spectral structure is characteristic of the chemical composition of the particle. The intensity of the light is directly proportional to the particle mass, i.e., the simple relationship

$$I = kd^3 \quad (6)$$

is true over the whole measurable particle size range, where  $I$  is the intensity of the light flash,  $k$  is a constant and  $d$  is the particle size. The light flash is converted into a electrical impulse by means of an optical electronic unit. The impulse amplitude is a measure of the particle mass or particle size, and the frequency of the impulses a measure of the number of particles.



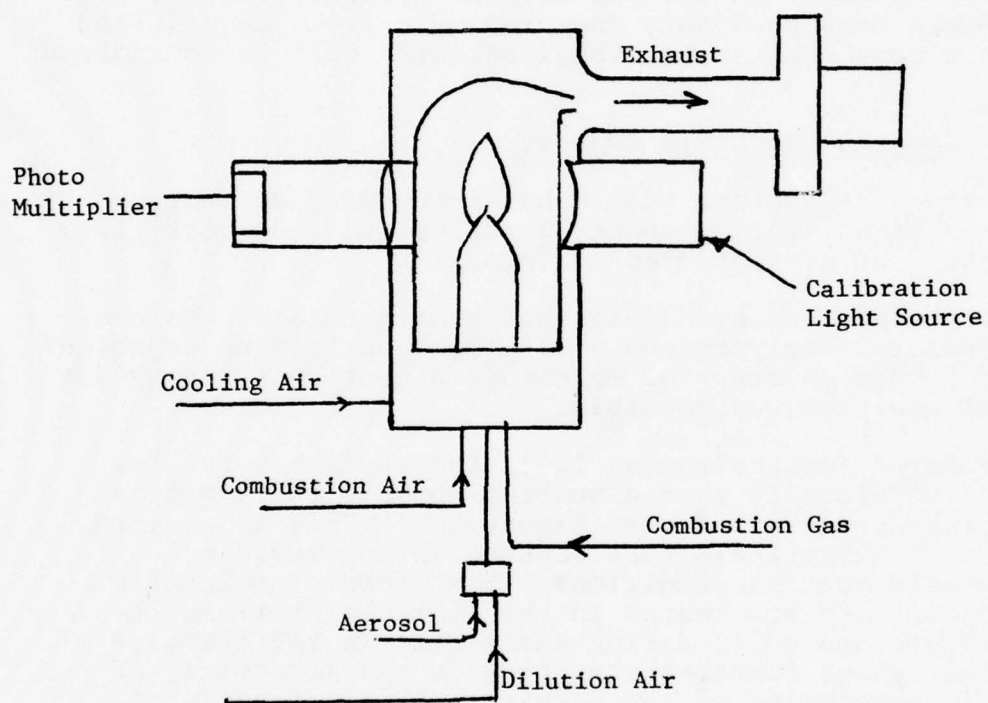


Figure 36: Sartorius Spectral-Particle Counter: Schematic Diagram.

The impulses are sorted into 10 adjustable size ranges by means of an amplitude analyzer. The sum frequency of the particles in 10 different particle size ranges are shown digitally on 10 counters. The particle concentration as well as the particle size distribution is therefore determined for a particular aerosol throughput. Furthermore, weight concentration in the range  $10^{-6}$  to  $10^{-2}$  g/m<sup>3</sup> can be measured and indicated in digits by means of an integrator.

The sensitivity of the instrument depends on the signal-noise ratio whereby the emission of the hydrogen flame and the noise of the photomultiplier determine the background. The use of a monochromator in the optical system reduces the flame background to such an extent that the sensitivity reaches the submicroscopic range, e.g., the smallest detectable particle diameter for NaCl is 0.02  $\mu$ m.

The logarithmic relationship between particle size and impulse amplitude, which is created electronically, makes it possible to record simultaneously on the 10 channels of the analyzer a particle size range of  $10^{-2}$  to 10 microns. By switching over to linear function, the fine distribution of any particular section of the whole particle size spectrum can be recorded on the 10 channels.

With the use of an appropriate monochromator the instrument can be set for an aerosol of a particular chemical composition so that particles of a different chemical nature cannot be recorded. On the other hand, it is possible to carry out qualitative analyses by varying the monochromator.

The dead time of the instrument and thereby the maximum resolvable particle concentration depends basically on the length of time the particle stays in the flame. This is approximately 5 msec. Since the coincidence of a number of particles in the flame is recorded as one big particle, only one particle at a time should be in the flame. Assuming a Poisson distribution of the aerosol and a counting error of approximately 10%, this means a maximum aerosol count rate without dilution of approximately 3,000 particles/min or 20 particles/cm<sup>3</sup>. The instrument contains a mixing unit with which the aerosol can be diluted 40 fold so that the undiluted aerosol count rate is in the order of  $10^3$  particles/min, i.e., 800 particles/cm<sup>3</sup>.

The spectral particle counter has an aerosol throughput of 200 cm<sup>3</sup>/min. The flow is determined by the 50 mm water column drop of pressure in the metal capillary which introduces the aerosol to be analyzed into the measuring chamber. Thermophoretic processes in the capillary, which is kept at a constant temperature, prevent particle separation on the capillary walls.

The instrument is provided with an isokinetic adaptor on the aerosol inlet. At the same time, it allows for the electronic control of dynamic and static pressure is provided at the aerosol inlet and thus control of the constancy of the aerosol throughput selected.

This instrument is useful for determining size and concentration of aerosols such as Na, Li, Sr, K, Pb, etc. which have a well characterized emission spectra. The instrument response has to be calibrated for each aerosol composition. It obviously cannot be used on a mixed aerosol of varying compositions.

Since the entire flame is the viewing volume, the coincidence errors are a big problem and limit the concentration of aerosol to less than 20 particles/cm<sup>3</sup> without dilution.

In conclusion, this instrument is useful for specific dilute aerosols having an invariant chemical composition and a well defined spectra. Its usefulness is therefore limited.

#### 2.2.5 Laser Interferometry

Farmer [26] presented a method for determination of particle size, number density, and velocity utilizing a laser interferometer. The analysis showed that if the interference fringe spacing is comparable to the particle size, its diameter can be estimated; and when the fringe spacing is much greater than the particle size, the number density can be measured. Farmer [27] reported observation of large particle signal generation by the passage of a particle through the interference pattern. Farmer [28] analyzed some parameters that can affect the sampling characteristics. The sample space depends directly on the scattering cross section of the particle. Hence, statistical measurements of size dependent properties must account for the fact that as the particle size increases, so does the space over which the population may be sampled.

Spectron Development Laboratories, Inc. [29] offers a custom system for size measurement with laser doppler anemometer.

The Particle Morphokinometer determines particle size and velocity through measurement of light scattered from an interference pattern generated by a lens set focusing a set of laser beams to a common center. The following discussion describes the principles behind how these measurements are accomplished.

A typical interferometer system for measuring particle velocity is shown in Figure 37. The region of measurement, called the probe volume or sample space, exists where the two beams cross and interfere. Light scattered out of this region

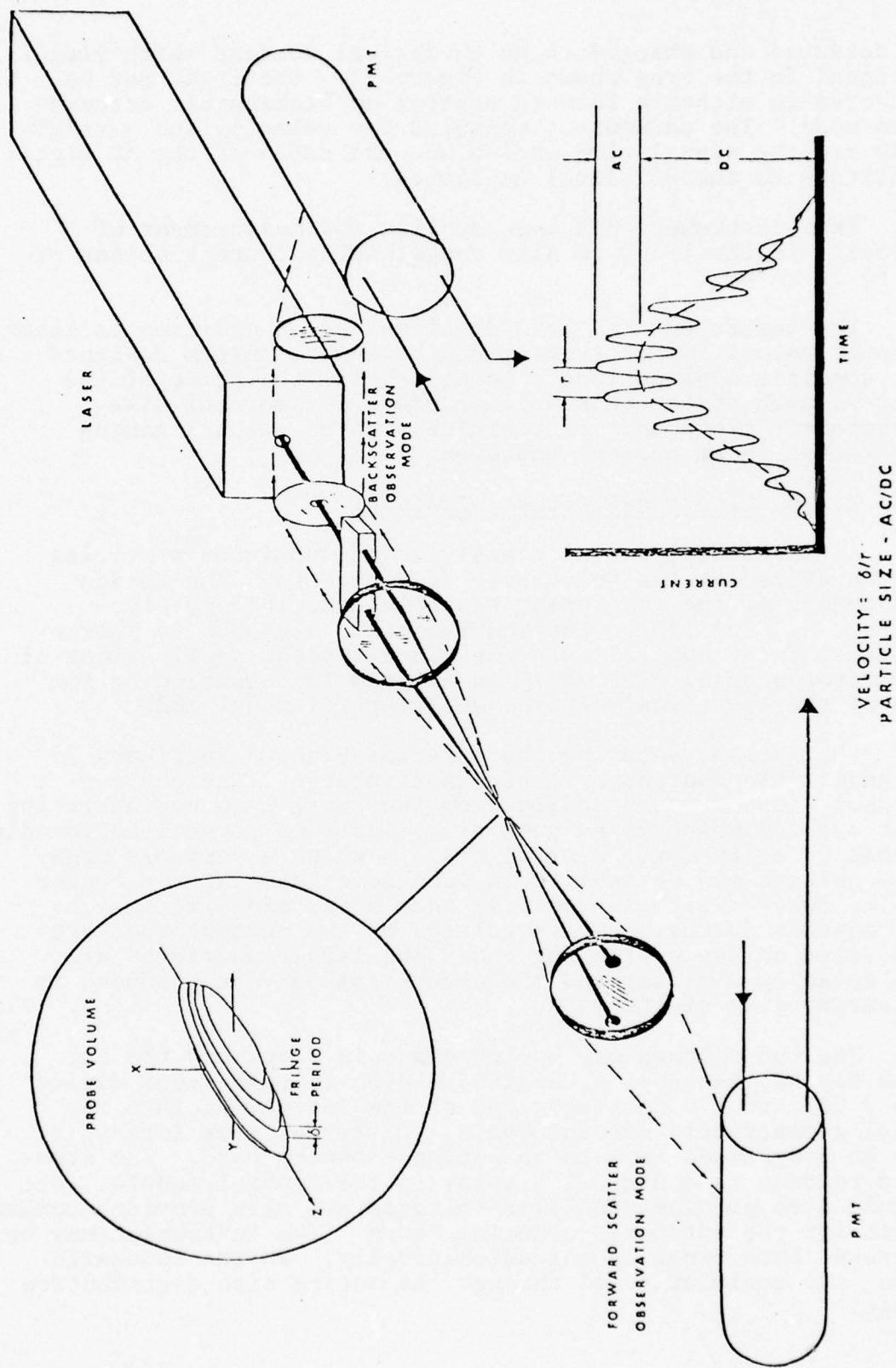


Figure 37: Standard Interferometer System for Velocity & Particle Size Measurement



is detected and changed to an electrical current which yields a signal of the type shown in Figure 37. The light may be detected in either a forward scatter or backscatter observation mode. The parameters measured for velocity and particle size are the signal time period and the ratio of the AC signal amplitude to the DC signal amplitude.

This instrument has been applied for measurement of aerosols in the 1-300  $\mu\text{m}$  size range having concentrations of up to 10  $\text{gm}/\text{m}^3$ .

The instrument has been developed as an addition to laser doppler velocitimeter systems and have been custom designed for specific applications. Consequently, the price of the unit is much higher than an instrument for aerosol size measurement alone and its usefulness as an aerosol sizing instrument is at present unproven.

#### 2.2.6 Electrical Aerosol Analyzer

An instrument for size analysis of submicron particles was developed at the University of Minnesota. The design and theory of the instrument has been described by Liu, Whitby, and Pui [30]. The instrument is marketed by Thermo-Systems, Inc. Sem [31] has described several applications of submicron aerosol size analysis and sample conditioning for use of the electrical aerosol analyzer TSI model 3030.

The aerosol entering the instrument shown in Figure 38 is positively charged by a corona discharge. The charged aerosol flows concentrically from the charger to the analyzing tube section which is an annular cylinder of aerosol surrounding a core of clean air. A metal rod, to which a variable negative voltage can be applied is located axially at the center of the tube. Particles smaller than a certain size, which corresponds to the negative voltage on the central rod, are deposited on the collecting rod. The larger particles are collected on a filter and the electrical current produced is measured by an electrometer.

The rod voltage may be increased in steps and the cut size may be changed. A total of eleven voltage steps divide the 0.0032 to 1.0  $\mu\text{m}$  size range of the instrument into ten equal geometrical size intervals. Different size intervals can be programmed in with an optional memory card. The standard readout is a digital display in the control module. The module also programs analyzer voltages and also provides command logic for the automatic counting modes. The instrument may be operated both manually and automatically. In the automatic mode, the analyzer steps through the entire size distribution range.

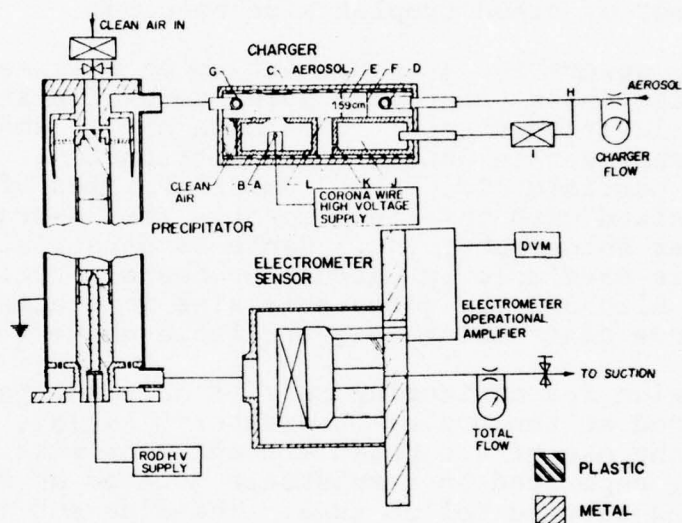


Figure 38: Electrical Aerosol Analyzer (TSI 3030)

The aerosol sample flow rate is 4 lpm. The surrounding sheath flow is 46 lpm and may be drawn from the aerosol source or other air environments such as room air. The aerosol concentration range is 1-1000  $\mu\text{g}/\text{m}^3$  which corresponds to about  $10\text{-}10^3$  particles/ $\text{cm}^3$  of 0.7  $\mu\text{m}$  in size.

### 2.3 Miscellaneous Instruments

Knollenburg [32] described the use of a photodiode array for imaging particles and processing the images to obtain their size distribution. The schematic diagram of the device is shown in Figure 39. PMS, Inc. markets two systems based on this principle. The first system is useful for aerosols in the 20-300  $\mu\text{m}$  and the second system is useful in the 300-450  $\mu\text{m}$  range. These instruments were developed for in-flight measurement of cloud droplet size spectrum.

Hotham [33] described an instrument which utilizes a pulsed ultraviolet laser to produce hologram of the aerosol droplets on a television screen. The image may be analyzed using an image analyzer to obtain size distribution. The minimum detectable particle size is 0.3  $\mu\text{m}$ . Particles of up to 1000  $\mu\text{m}$  may be sized with this instrument. The instrument is marketed by Laser Holography, Inc., Santa Barbara, California. The instrument is particularly useful for characterizing coarse spray systems. Although the detectable size is claimed to be 0.3  $\mu\text{m}$ , we believe that sizing to be reliable above 2  $\mu\text{m}$ .

A novel device for monitoring mass of airborne particles is being developed at the Dudley Observatory, Albany, N.Y. [34]. It is based on the use of a tapered element as a mass sensor. Particles may be deposited on a substrate mounted on the closed, narrow end of the tapered hollow tube. The wide end is firmly mounted. The free end is set into oscillation. A feedback system maintains the oscillation whose natural frequency will change in relation to mass deposited on the substrate. The sensitivity of the instrument can be an order of magnitude higher than the commercial quartz crystal monitors (QCM). Several other advantages over the QCM's are claimed. Uniformity of deposition necessary for proper functioning of the QCM may not be necessary for the tapered element device. The instrument is presently in a development stage but is potentially applicable to a wide range of applications.

A major limitation in the commercial aerosol instruments available today is that most of the instruments are capable of handling only relatively dilute aerosols of concentrations up to  $10^4$  particles/ $\text{cm}^3$ . Dilutions by a factor of up to  $1:10^3$  may be needed for dense aerosols such as smokes. The commercial diluters using conventional dilution techniques are capable of only  $1:10^2$  dilution without significant errors. Progress in developing measurement techniques for concentrated aerosols as well as reliable dilution system is urgently needed.

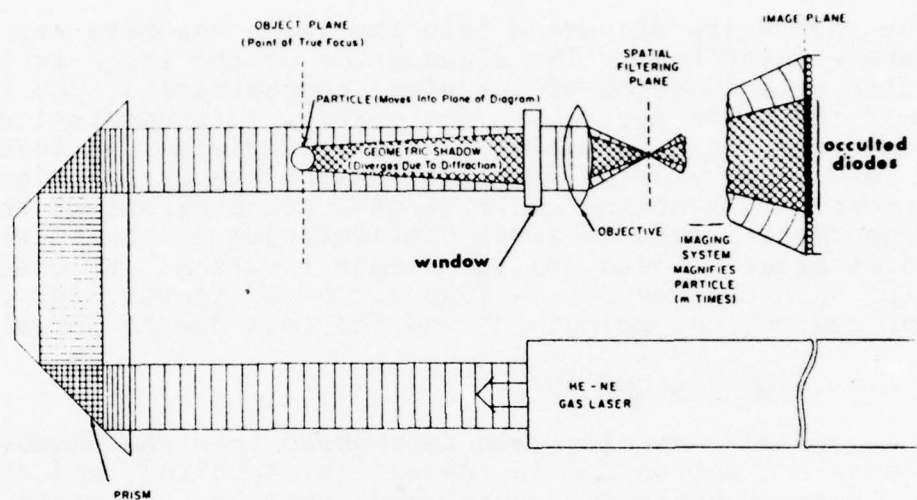


Figure 39: Schematic of Imaging System Designed to Size Droplets with Airborne Equipment



### 3. AEROSOL INSTRUMENTATION FOR THE CHAMBER AEROSOL STUDIES

The Munitions Applications Branch at Edgewood Arsenal is conducting aerosol studies in two large cylindrical chambers. The aerosol systems to be studied may be grouped into two types: smokes and chemical agents. The smokes are composed of hygroscopic liquid particles in the 0.1 to 1  $\mu\text{m}$  size range. The concentration of the smokes may reach up to  $10^6$  particles/ $\text{cm}^3$ . The solid agents range from a few microns to 100  $\mu\text{m}$  in size and their concentrations may be as high as  $10^3$  particles/ $\text{cm}^3$ .

#### 3.1 Studies with Smoke Aerosol

The smokes are dispersed into the large chambers and they attenuate a laser beam. The attenuation of the laser is to be studied as a function of the size, concentration, and the nature of the smoke aerosols. The aerosol instrumentation system should provide information regarding the size and loading of the aerosol to provide correlation with the attenuation measurement. In sampling smoke aerosol for monitoring, the following factors must be given consideration as potential sources of error. These are (a) sample location, (b) coagulation, (c) size changes due to evaporation or growth, (d) relevance of measurement principle, and (e) loss due to deposition.

##### 3.1.1 Sample Location

The aerosol particles when introduced into the chamber are in constant motion due to convection, settling, and diffusion. For the micron and submicron particles, particle diffusion is dominant over gravitational or inertial settling. Particles of 0.1 and 1  $\mu\text{m}$  size have mean Brownian displacements of  $2.95 \times 10^{-3}$  cm/sec and  $5.9 \times 10^{-4}$  cm/sec, respectively. In comparison, the settling velocities of these particles are  $8.64 \times 10^{-5}$  cm/sec and  $1.28 \times 10^{-2}$  cm/sec, respectively. Convective motion and the Brownian motion of the particles tend to equalize the aerosol concentration in the chamber. However, over several hours a decay of the aerosol concentration may take place due to deposition by diffusion.

In the smoke study, best results will be expected by monitoring the aerosol characteristics in or near the laser beam.

##### 3.1.2 Coagulation

All particles in an aerosol cloud are in constant motion due to Brownian, convective, and gravitational forces. When two particles come into contact with each other during their motion, they can adhere and then act as a single entity. This process occurs throughout the cloud and is called the coagulation of the aerosol.

The extent of coagulation is dependent on the probability of collisions between the particles. Their Brownian motion is a random process and the probability of the collisions is dependent on the mean particle velocity. This is a function of the particle size and it increases as the size of the particle decreases.

Convection due to thermal or hydrodynamic forces also causes a relative motion of the particles which contributes to coagulation.

In gravitational field, the larger particles settle faster than the smaller ones, and during settling they come into contact with them contributing to coagulation.

The simplest form of theory was developed by Smoluchowski. Considering the aerosol to be monodisperse and the coagulation to occur by Brownian motion alone, he derived the following equation for the rate of decrease of particle number concentration

$$\frac{dn}{dt} = -K_0 n^2 \quad (6)$$

A useful form of this equation is obtained by integration of equation

$$\frac{1}{n} - \frac{1}{n_0} = K_0 t \quad (7)$$

where  $n$  ( $\#/cm^3$ ) is the number concentration at time  $t$ ,  $n_0$  is number concentration at time  $t = 0$ , and  $K_0$  is the coagulation constant.

Equation 7 indicates that a plot of  $1/n$  versus  $t$  is a straight line with a slope  $= K_0$ .

It has been established by experiments that this straight line relationship holds until the concentration drops to quite a low value (one thirtieth of the initial) for monodispersed aerosols.

For polydispersed aerosols, the Smoluchowski theory holds within experimental error, at least in the initial stages. The value of  $K_0$  is usually higher than predicted by equation 7 due to polydispersity. Ranade [35] showed that a polydispersity factor could be used to correlate the experimental coagulation coefficient to that predicted by theory for a monodisperse system.

For a  $1 \mu m$  aerosol, the coagulation coefficient is  $3.44 \times 10^{-10} cm^3/sec$ . If the initial aerosol contains  $10^6$  particles/ $cm^3$ , the concentration is halved in about 5 min.

For a  $0.1 \mu\text{m}$  aerosol, the coagulation coefficient is  $4.18 \times 10^{-10}$  and the concentration will be halved to  $5 \times 10^5$  particles/cm<sup>2</sup> in about 4 minutes.

The size, number concentration, and related properties of the aerosol are changed due to coagulation. In sampling, a dense aerosol dilution of the sample by clean air may be used to prevent coagulation from proceeding in the sampling lines. As pointed out later, most instruments would require dilution for proper operation and this dilution would also serve to stop coagulation in the sampling lines.

### 3.1.3 Relevance of Measurement Principle

The smoke study is aimed at correlated attenuation and other optical properties of a laser by the smoke aerosols. The attenuation and scattering of the particles is dependent on size and number concentration of the aerosols. The sizing method employed should be based on a principle of measurement similar to the studied phenomena. Use of optical sizing methods such as scattering is to be preferred. The number density and mass concentration must also be determined. Commercial aerosol light scattering counters can be used to measure the size distribution and number density of the aerosol. The expected smoke aerosol number density is, however, several orders of magnitude higher than the capabilities of most of the monitors. Dilution of the aerosol is therefore necessary.

Mass measurement may be accomplished with the help of commercial mass monitors after dilution.

### 3.1.4 Evaporation and Growth of Aerosol Droplets

The size distribution of a hygroscopic aerosol is dependent on the relative humidity of its environment. In sampling such aerosols for size analysis, growth or evaporation which may change the particle size must be minimized. If a dilution of the sample is required, the humidity of the original aerosol should be maintained. This may be done conveniently, by using a filtered stream of the aerosol sample for dilution of the sample stream. The measurement method should preferably be based on the size measurement in flight rather than deposition of a surface. Optical sizing techniques such as light scattering are to be preferred.

### 3.1.5 Deposition of Aerosols

Deposition loss of particles in sampling lines can affect the size distribution as measured by the sensor and also hinder sampling efficiency of continuous monitors. For micron and submicron aerosols, the deposition may take place by diffusion. Ranade [36,37,38] developed a sampling probe for quantitative sampling of aerosols. Such a probe can serve the purpose of dilution as well as sample preservation.



### 3.1.6 Recommended Sampling Set-Up for the Smoke Studies

Based on the preceeding discussion, the sample system must be designed so that a representative sample may be obtained from the chamber with minimum modification of the sample. A schematic of the sampling system is shown in Figure 40.

The sample should be drawn from the center of the chamber and at other points along the laser path. Another stream of the sample is drawn from the chamber, filtered with a glass wool filter and returned to the sampling tube to provide a transpiration dilution sheath. The total sample will be drawn through the mass monitor or size analyzer as required.

### 3.1.7 Studies with Solid Agents

Solid agents to be dispersed into the chamber will be in the 1-100  $\mu\text{m}$  size range and up to  $10^3$  particles/ $\text{cm}^3$  may be present. The properties to be studied are related to particle motion, dispersion, and deposition. The size should be based on aerodynamic particle sizing. Although the size of the airborne particulates may be as high as 100  $\mu\text{m}$  particles, such large particles will remain suspended only for a short time before settling. In Table 11, the terminal velocity of particles in air is shown for different particle sizes. Time for settling of particles from the top of a 15 ft chamber are shown in Table 11. Vigorous stirring may reduce the deposition losses, somewhat.

The size classification of the solid agents should preferably be based on inertial principle such as cascade impactors. The effects of shape and density are accounted for by measuring the aerodynamic diameter which is the diameter of a sphere of unit density having the same settling velocity of the particles measured.

The inertial classification based on aerodynamic diameter is obtained only by using a cascade impactor. Unfortunately, only one size analysis instrument based on impactors is available. In addition, losses due to particle bounce can produce significant errors in collection by the cascade impactor for particles above 10-15  $\mu\text{m}$ .

The sampling line losses of the particles due to settling also create serious problems. This may be overcome by using the transpiration probe described by Ranade [36]. The transpiration sampling probe utilizes a radial inward flow of clean air from the walls of the probe. This sheath prevents the particles from reaching the probe walls where they may be deposited. Particles up to 70  $\mu\text{m}$  have been successfully transported through such a sampling probe.



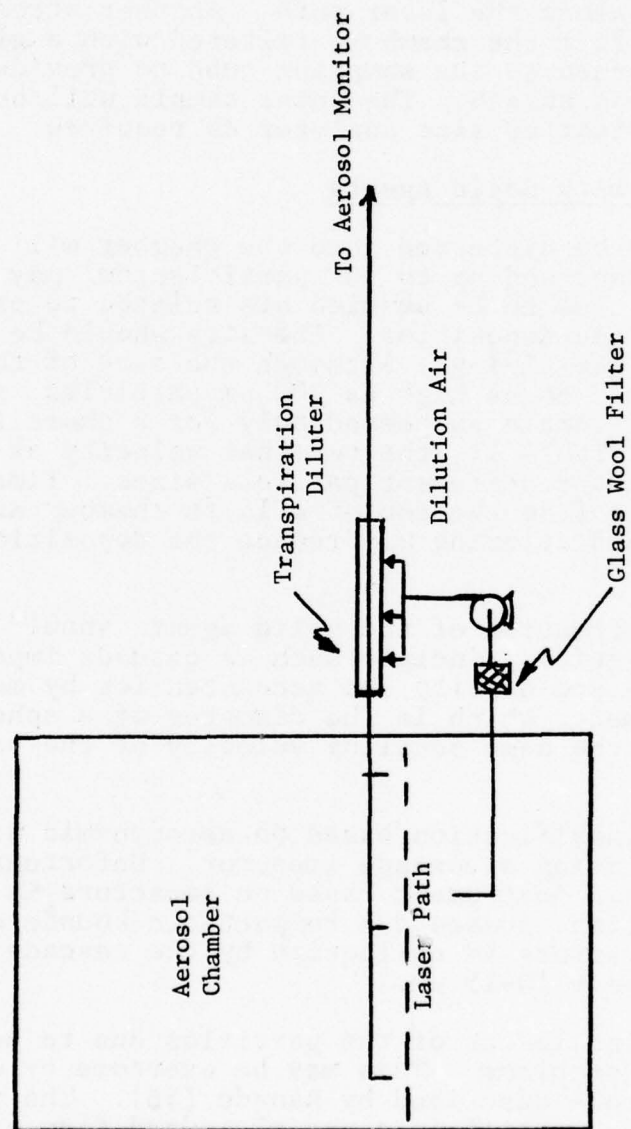


Figure 40: Sampling arrangement for smoke studies.

Table 11

TIME FOR UNIT DENSITY PARTICLES SETTLING FROM 15'  
HEIGHT TO THE FLOOR OR THE CHAMBER

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<u>Particle Size (<math>\mu\text{m}</math>)</u>	<u>Settling Velocity cm/sec</u>	<u>Time (min)</u>
1	$3.5 \times 10^{-3}$	2100
5	$7.7 \times 10^{-2}$	970
10	$3.06 \times 10^{-1}$	42
20	1.21	6.2
40	4.75	1.58
60	10.2	1.03
70	13.6	0.56
100	24.8	0.03

---

Commercial size analysis devices are also limited to particles below 30  $\mu\text{m}$  since large particles are usually suspended for only a small period. Also the sampling efficiencies of the instrument drop off to a very low value for larger particles.

The commercial instruments capable of covering the large size range at least partially are based primarily on light scattering measurement. Only one device using a piezoelectric microbalance is available for size measurement on aerodynamic classification. If the optical sizing techniques are to be used, variations of shape, density, and optical properties of the agent will require calibration whenever the aerosol size characteristics or composition is changed.

### 3.2 Aerosol Instruments Applicable to the Chamber Studies

The instrument systems for the aerosol studies in the large chambers are required to measure size distribution and concentration over a wide range. The choice of instruments for mass measurement is described below.

#### 3.2.1 Mass Measurement

The direct mass measurement instruments based on the piezo electric microbalance and the  $\beta$ -ray attenuation principle are preferable over aerosol photometers which do not respond directly to mass. The list of mass monitors is presented in Table 12. These are identified as M-1, M-2, etc. for the following discussions.

M-1 is the Celesco IBC (Berkeley) mass monitor based on quartz crystal microbalance (QCM). M-2 is also a Celesco IBC (Berkeley) instrument which utilizes the QCM principle and a cascade impactor to obtain size distribution based on mass as well as the mass concentration. M-3 is TSI mass monitor based on QCM principle. M-4 is a mass monitor made by GCA, Inc., and is based on  $\beta$ -ray attenuation by a sample collected on an impactor. The M-5 mass monitor is also made by GCA, Inc., and is based on  $\beta$ -ray attenuation by aerosols collected on a filter. The performance of these instruments were discussed in Sections 2.1.5 and 2.1.6.

The applicability of the instruments as mass monitors to the smoke studies varies depending on the size range capabilities. Instruments M-1 and M-4 use impactors for collection. The smoke which is submicron in size may be fully collected by these instruments. M-2 incorporates an impactor stage for collection of particles below 0.3  $\mu\text{m}$  and down to 0.05  $\mu\text{m}$ , using reduced pressure. As mentioned in Section 2.1.6, the evaporation of aqueous hygroscopic particles under the reduced pressure will prevent determination of the true mass of the aerosol in this size range. The added advantage of

Table 12

## MASS MONITORS APPLICABLE TO CHAMBER STUDIES

No	Manufacturer	Model No.	Principle	Size Range ( $\mu\text{m}$ )	Concentration Range ( $\mu\text{gm}/\text{m}^3$ )	Required Dilution
M-1	IBC (Berkley)	PM-40	QCM	0.3 - 20	$2 \times 10^3$	$1:10^3$
M-2	IBC (Berkley)	C-1000	QCM	0.05 - 20	$10^2 - 6 \times 10^4$	$1:10^2$
M-3	TSI, Inc.	3205B	QCM	0.01 - 20	$<2 \times 10^5$	1:10
M-4	GCA Inc.	RDM-101	3205B	0.5 - 20	$<1.5 \times 10^5$	1:10
M-5	GCA Inc.	PMM-1	PMM-1	0.5 - 20	$<1 \times 10^6$	--



size measurement to 0.3  $\mu\text{m}$  is however, an attractive one, especially for the larger solid agent aerosols.

The M-3 monitor uses electrostatic collectors with excellent collection efficiency down to 0.05  $\mu\text{m}$ . The dilution of the sample required can also be reasonably achieved. The M-5 monitor uses a glass fiber filter for particle collection. Inhomogeneities of the filter medium may cause significant errors. Other sources of errors include changes in chemical composition as discussed in Section 2.1.6 and the loss of volatile matter prior to the mass determination.

All of these monitors are applicable to the solid agent assessment. However, the upper size measured is limited by inlet efficiencies to  $\sim 20\text{-}30\ \mu\text{m}$ .

### 3.2.2 Size Analyzers Applicable to Chamber Studies

The size analyzers of particle size measurement for both aerosol systems are shown in Table 13. These are identified by numbers S-1, S-2, S-3, S-4, S-5, S-6, S-7, and S-8.

S-1 is a laser light scattering device using a 0.94  $\mu\text{m}$  laser as a light source. The size range may be selected from several size ranges with a upper and lower size ratio of 15:1 in one instrument. The instrument is described in detail in Section 2.2.2. The applicability may be limited to either the smoke or the chamber studies.

S-2 is another laser light scattering device that is primarily applicable for the smoke studies. The particle size range is divided into four overlapping sub-ranges, each range being divided into 15 channels. The size and other operating data displayed on a cathode ray tube, and a digital display of a selectable channel. A printer can also be used to obtain a hard copy of the data.

The next three devices, S-3, S-4, and S-5 are all commercial light scattering photometers using conventional optics. The Royco 225 and the Tech Ecology 208 are both forward light scattering instruments. The Climet 208 instrument uses a unique light collection scheme to collect the light scattered over a wide angle range. These instruments have been discussed in detail in Section 2.2.1. The capabilities of these instruments are comparable to each other. The Climet 208 potentially is the better of these instruments since it gathers light over a wide angel range. The dilution required for these instruments is rather high for smoke aerosols. These instruments are applicable mostly to solid agents. The S-6 is the TSI aerosol analyzer and is useful only for measurements with smoke aerosols. Dilution requirement is rather high. The resolution of the instrument is good. The size measurement is based on electrical mobility and the measurement

Table 13  
SIZE ANALYZERS FOR CHAMBER STUDIES

No.	Manufacturer	Model	Principle	Size Range ( $\mu\text{m}$ )	Concentration Range (particles/cm <sup>3</sup> )	Required Maximum Dilution
S-1	ESC	PILS-IV	Near Forward	0.2 - 3	$5 \times 10^5$	--
S-2	PMS	ASAS	Active Laser Cavity Scattering	0.1 - 5	$10^4$	$1:10^2$
S-3	Royco	225/518	Near Forward	0.3 - >30	$10^2$	$1:10^3$
S-4	Climet	208	15 - 150° Total Scatter	0.3 - >10	$10^2$	$1:10^3$
S-5	Tech Ecology	208	Near Forward	0.5 - >10	10	$1:10^3$
S-6	TSI	3030	Electrical Mobility	0.003 - 1.0	$1000 \mu\text{g}/\text{m}^3$	$1:10^3$
S-7	IBC	C-1000	Cascade Impactor + QCM	0.05 - 20	$60,000 \mu\text{g}/\text{m}^3$	$1:10^2$
S-8	Sartorius	SM-167-16	Scintillation	0.01 - 100	20	$1:10^5$

principle is not directly related to the aerosol properties under study. It is discussed fully in Section 2.2.6.

S-7 is the same as the M-2 mass monitor. It uses QCM balance to measure masses of particles in several impactor stages. The applicability of the instrument to the smoke studies is questionable due to poor resolution and use of reduced pressure for collection of particles in the 0.05 to 0.3  $\mu\text{m}$  range which may cause evaporation of these particles.

S-8 is based on spectroscopic determination of chemical species as discussed in Section 2.2.6. Its applicability is limited to a very few substances. Due to the large flame which is the sensing zone, the concentration of particles that may be processed is rather limited.

All of these instruments have a sufficiently fast response and are fully automated so that a hard copy of the data is obtainable from digital printers. The flow rates required are low enough to cause little change of chamber contents by long term use (over several hours). The S-7 instrument will require cleaning of the crystals.

Other instruments are all chosen so that continuous sampling and automatic print record of the data are obtained. These instruments may be operated on non-continuous mode if desired.

### 3.3 Cost and Benefit Evaluation

Selection of instrumentation for the chamber studies may be conducted using a cost and benefit evaluation. The benefits of an instrument may be obtained by grading required benefits and evaluating instrument performance against these. Of the possible mass monitors and size analyzers in Tables 12 and 13, a few were chosen to obtain a series of combinations of instruments that may be utilized for the chamber studies. In Table 14, a list of the combinations used in the example is given. These particular instruments were selected from the list in Tables 12 and 13 as the most applicable for the chamber studies.

The combination C-1 represents only the IBC Celesco impactor (#C 1000) which is equipped with the quartz crystal microbalances in every stage. The applicability of the instrument to smoke studies is questionable.

In the list shown in Table 14, a single mass monitor was considered adequate to work effectively on both the smoke and solid agents studies. The size analyzers are listed such that the first instrument applicable is for smoke studies and the second instrument is for solid agents in the list.

Table 14

## COMBINATION SYSTEMS FOR CHAMBER STUDIES

<u>Combination No.</u>	<u>Mass Monitor</u>	<u>Size Analyzer</u>
C-1	M-2	S <sub>7</sub> (M <sub>2</sub> )
C-2	M-3	S-1 (Smoke), S-1
C-3	M-3	S-1 (Smoke), S-3
C-4	M-3	S-1, S-4
C-5	M-3	S-1, S-5
C-6	M-3	S-2, S-1
C-7	M-3	S-2, S-3
C-8	M-3	S-2, S-4
C-9	M-3	S-2, S-5
C-10	M-3	S-6, S-1
C-11	M-3	S-6, S-3
C-12	M-3	S-6, S-4
C-13	M-3	S-6, S-5

S-7 and M-2 are identical instruments.



The instruments considered in the evaluation are presented in Table 15. The instruments are identified by their model numbers and manufacturers names and addresses. Auxiliary instruments such as printers and recorders are also included. Cost information was obtained from manufacturers data. Additional information is found in Tables 12 and 13, and in the manufacturers brochures supplied separately to the Project Officer.

The dilution system is not shown separately as it will be common to all the systems. The transpiration dilution system is not commercially available. It is estimated that a satisfactory dilution system could be obtained for under \$10,000.

Thirteen systems were composed from the possible combinations of mass monitors and size analyzers which could acceptably meet the technical requirements of size and mass determination.

The evaluation of these alternative systems is based on a benefit/cost index for each system under consideration. Benefits are estimated from relevance numbers accumulated for the instruments used to make-up the system. Costs are represented solely by acquisition price. This analysis and the results are now discussed.

#### 3.3.1 Systems Benefits

A list of factors representing the advantages or disadvantages among the instruments was constructed. These factors were then rated on an exponential scale in accordance with their importance\*. The scores for each factor were then normalized to obtain the corresponding relevance numbers. The relevance numbers represent the relative importance among the factors under consideration. These results are presented in Table 16.

In a similar manner, each of the applicable instruments were rated with respect to the nine factors (Table 17)\*\*. Again the ratings were made on an exponential scale and then normalized to obtain relevance numbers (Table 18) for mass monitors and size analyzers separately (since their benefits relate to different functions).

The relevance numbers for the factors and for the instruments were used to determine the benefits of the various instruments. Referring to Tables 19 and 20, the "TOTAL" for

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\* Linear ratings are easier to implement but do not adequately separate important from unimportant items. Therefore, the factors were originally placed on a scale from 1 to 5, then transformed to 1, 2, 4, 8, and 16 correspondingly.

\*\* Also rated on exponential scale, as in footnote \*.

Table 15

## COST INFORMATION OF SELECTED INSTRUMENTS

<u>Instrument</u>	<u>Manufacturer</u>	<u>Model No.</u>	<u>Auxiliary Equipment</u>	<u>Cost, \$</u>
M-2, S-7	IBC-Berkley, Irvine, CA	C-1000	--	12,000
M-3	TSI-Inc. 2500 Cleveland Ave. North St. Paul, Minn. 55113 612/633-0500	3205B	--	4,600
S-1	Environmental Systems Corporation P.O. Box 2525 Knoxville, TE 37901 615/637-4741	PILS IV	--	36,600
S-2	Particle Measuring Systems 1855 S. 57th Ct. Boulder, CO 80301 303/443-7100	ASAS-300A	HP 5055A Printer	16,000
S-3	Royco Instrument Co. 141 Jefferson Drive Menlo Park, CA 94025 415/325-7811	225/518		9,600
S-4	Climet Instrument Co. 1620 West Colton Ave. Redlands, CA 92373 714/993-2788	CI-208	CI-235A Printer	9,600
S-5	Tech. Ecology, Inc. 645 S. Mary Ave. Sunnyvale, CA 94086 408/736-6950	208	Option 6: Digital Printer	5,500
S-6	TSI-Inc. (same as for M-3)	3030		10,700

Table 16  
RELEVANCE NUMBERS FOR BENEFIT FACTORS

<u>Factor</u>	<u>Exponential Rating</u>	<u>Relevance Number</u>
Speed	2	0.05
Accuracy	4	0.10
Automation	8	0.21
Reliability	2	0.05
Portability	2	0.05
Versatility	2	0.05
Decontamination Probe losses	1	0.03
Applicability of Principle	2	0.05
Operator Time	16	0.41

Table 17

EXPONENTIAL RATINGS OF INSTRUMENTS WITH  
RESPECT TO BENEFIT FACTORS

Factor	Mass Monitors		Size Analyzers					
	M-2	M-3	S-1	S-2	S-3	S-4	S-5	S-6
Speed	1	1	4	8	2	2	2	2
Accuracy	1	1	16	2	1	4	1	16
Automation	2	1	1	2	2	2	2	2
Reliability	1	1	16	2	1	2	1	16
Portability	1	1	2	2	2	2	2	1
Versatility	4	1	1	1	4	4	4	1
Decontamination Probe Losses	1	1	8	1	1	1	1	1
Applicability of Principle	1	1	16	2	4	4	4	1
Operator Time	1	1	1	8	2	2	2	2



Table 18

## RELEVANCE NUMBERS FOR INSTRUMENTS

Factor	Mass Monitors			Size Analyzers				
	M-2	M-3	S-1	S-2	S-3	S-4	S-5	S-6
Speed	0.50	0.50	0.19	0.38	0.10	0.10	0.10	0.10
Accuracy	0.50	0.50	0.36	0.05	0.02	0.09	0.02	0.36
Automation	0.67	0.33	0.08	0.17	0.17	0.17	0.17	0.17
Reliability	0.50	0.50	0.38	0.05	0.02	0.05	0.02	0.38
Portability	0.50	0.50	0.17	0.17	0.17	0.17	0.17	0.08
Versatility	0.80	0.20	0.05	0.05	0.21	0.21	0.21	0.05
Decontamination Probe Losses	0.50	0.50	0.53	0.07	0.07	0.07	0.07	0.07
Applicability of Principle	0.50	0.50	0.25	0.03	0.06	0.06	0.06	0.02
Operator Time	0.50	0.50	0.05	0.42	0.11	0.11	0.11	0.11

Table 19  
BENEFITS OF MASS MONITORS

<u>Factor</u>	<u>Weight</u>	<u>Benefits of Mass Monitors</u>	
		M-2	M-3
Speed	5.0	2.5	2.5
Accuracy	10.0	5.0	5.0
Automation	21.0	14.1	6.9
Reliability	5.0	2.5	2.5
Portability	5.0	2.5	2.5
Versatility	5.0	4.0	1.0
Decontamination Probe Losses	3.0	1.5	1.5
Applicability of Principle	5.0	2.5	2.5
Operator Time	41.0	20.5	20.5
Total		55.1	44.9
Benefit		110.2	89.8

Table 20

## BENEFIT OF SIZE ANALYZERS

Factor	Weight	Benefits of Size Analyzers					
		S-1	S-2	S-3	S-4	S-5	S-6
Speed	5.0	1.0	1.9	0.5	0.5	0.5	0.5
Accuracy	10.0	3.6	0.5	0.2	0.9	0.2	3.6
Automation	21.0	1.7	3.5	3.5	3.5	3.5	3.5
Reliability	5.0	1.9	0.2	0.1	0.2	0.1	1.9
Portability	5.0	0.8	0.8	0.8	0.8	0.8	0.4
Versatility	5.0	0.3	0.3	1.1	1.1	1.1	0.3
Decontamination Probe Losses	3.0	1.6	0.2	0.2	0.2	0.2	0.2
Applicability of Principle	5.0	1.3	0.2	0.3	0.3	0.3	0.1
Operator Time	41.0	2.2	17.3	4.3	4.3	4.3	4.3
Total		14.4	24.9	11.0	11.8	11.0	14.8
Benefit		100.8	174.3	77.0	82.6	77.0	103.6

each instrument was obtained by multiplying the relevance number (of that instrument with respect to a factor) by the weight of the corresponding factor, then summing over all the factors\*. By this operation, the ratings for each instrument have been weighted according to the importance of each factor\*\*.

It should be noted that the "TOTAL" is distributed by the number of instruments. For example, with two mass monitors their sum of "TOTAL's" equal 100. Moreover, if they had equivalent benefits, then the values would each be 50 and 50. Similarly for the seven size analyzers, if each had equivalent benefits, the values would each be 14.3 (i.e., one-seventh of 100).

To obtain the "BENEFIT" of each instrument, the "TOTAL" is multiplied by the number of instruments (2 for mass monitors and 7 for size analyzers). Now, instruments with equivalent benefits would have a value of 100. Moreover, the benefits from any of the instruments are on the same scale. This is necessary to avoid a dependence on the number of instruments under consideration.

For the determination of system benefits, a contribution is made by the instrument used in each of four missions, designated as follows:

Mass-A: mass determination of smokes,

Mass-B: mass determination of chemical agents,

Size-A: size determination of smokes, and

Size-B: size determination of chemical agents.

Since each mission is equally important, one-fourth of the benefit for the appropriate instrument is accumulated for each mission\*\*\*. The "SYSTEM BENEFIT" is the total benefit

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\* The weights of the factors are simply the relevance numbers (in Table 15 multiplied by 100). This is done to retain significance to the first decimal place.

\*\* Looking at Table 18, we note that speed has a weight of 5.0 (the relevance number 0.05 times 100). For the mass monitor M-2, we get  $5 \times 0.5 = 2.5$  where the value of 2.5 represents the benefit contribution of "speed" for that instrument. The "TOTAL" represents the total of such contributions.

\*\*\* For example, in Table 20 under system C-1, we get the benefit of using instruments M-2 and S-9 (the IBC C-1000 for both mass and size determinations), thus,  $27.6 = (\frac{1}{4})(55.1)$  and  $21.0 = (\frac{1}{4})(84.0)$ .



towards completing all four missions resulting from the use of the respective instruments. These results are presented in Table 21.

### 3.3.2 Acquisition Costs

The acquisition cost of each system represents the total price of the necessary instruments and accessories to meet the requirements of the four missions previously discussed. The accumulation of these costs for each of the thirteen systems under consideration are summarized in Table 22 on the following page\*.

### 3.3.3 Benefit/Cost Index

For each system, the ratio of system benefits to the acquisition cost was computed. In this way, the optimum benefit/cost index is selected by the highest benefit value per unit of cost. These results are presented in Table 23.

The three systems with the highest benefit/cost index are as follows:

<u>System</u>	<u>Instruments</u>
C-1	IBC C-1000
C-13	TSI 3205, TSI 3030 and TechEcology 208
C-9	TSI 3205, PMS ASAS and TechEcology 208

Systems C-12, C-11, C-7, and C-8 follow in a second group with nearly equivalent benefits per unit of cost (3.6-3.7). Systems C-6, C-5, C-3, C-4, and C-10 follow as a third nearly equivalent group (1.8-2.0). System C-2 was the most costly arrangement and reflected an unattractive benefit to cost ratio of 1.2.

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\* Referring to Table 21: System C-1 represents one instrument costing \$12,000. System C-2 represents three instruments: the mass monitor M-3 and two size analyzers (both S-1), costing \$4,600, \$36,000, and \$36,000, respectively, (rounded to \$100's). Thus, system C-2 costs \$77,880 in total.

Table 21

## SYSTEM BENEFITS

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<u>System</u>	<u>Mass of Smokes</u>	<u>Mass of Chemical Agents</u>	<u>Size of Smokes</u>	<u>Size of Chemical Agents</u>	<u>System Benefits</u>
C-1	27.6	27.6	21.0	21.0	97.2
C-2	22.4	22.4	25.2	25.2	95.2
C-3	22.4	22.4	25.2	19.2	89.2
C-4	22.4	22.4	25.2	20.6	90.6
C-6	22.4	22.4	43.6	25.2	113.6
C-7	22.4	22.4	43.6	19.2	107.6
C-8	22.4	22.4	43.6	20.6	109.0
C-9	22.4	22.4	43.6	19.2	107.6
C-10	22.4	22.4	25.9	25.2	95.9
C-11	22.4	22.4	25.9	19.2	89.9
C-12	22.4	22.4	25.9	20.6	91.3
C-13	22.4	22.4	25.9	19.2	89.9

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Table 22

## SYSTEM COSTS

Acquisition Costs (Thousands of Dollars)

<u>System</u>	<u>First Instrument</u>	<u>Second Instrument</u>	<u>Third Instrument</u>	<u>Total</u>
C-1	12.0	N/A	N/A	12.0
C-2	4.6	36.6	36.6	77.8
C-3	4.6	36.6	9.6	50.8
C-4	4.6	36.6	9.6	50.8
C-5	4.6	36.6	5.5	46.7
C-6	4.6	16.0	36.6	57.2
C-7	4.6	16.0	9.6	30.2
C-8	4.6	16.0	9.6	30.2
C-9	4.6	16.0	5.5	26.1
C-10	4.6	10.7	36.6	51.9
C-11	4.6	10.7	9.6	24.9
C-12	4.6	10.7	9.6	24.9
C-13	4.6	10.7	5.5	20.8

Table 23  
BENEFIT/COST INDEX

<u>System</u>	<u>Benefits</u>	<u>Cost</u>	<u>Benefit/Cost</u>
C-1	97.2	12.0	8.1
C-2	95.2	77.8	1.2
C-3	89.2	50.8	1.8
C-4	90.6	50.8	1.8
C-5	89.2	46.7	1.9
C-6	113.6	57.2	2.0
C-7	107.6	30.2	3.6
C-8	109.0	30.2	3.6
C-9	107.6	26.1	4.1
C-10	95.9	51.9	1.8
C-11	89.9	24.9	3.6
C-12	91.3	24.9	3.7
C-13	89.9	20.8	4.3



An important observation is that costs have emerged as the dominant variable. This is reflected in Table 24 where it is noted that high benefit-to-costs correspond to low-cost systems, and low benefit-to-costs to high-cost systems. Thus, the added benefits (within consideration) over the entire range of systems are not cost-justified. That is, the high cost systems do not reflect sufficient benefits for their justifications. To further discriminate the advantages of the various systems, a total cost analysis would be needed. This would involve converting the benefit factors in time and dollars and then selecting the lowest cost system including both acquisition and operating costs over a determined time frame.

It is recommended that in choosing the final system, the applicability of the instrument to the property under study should be considered of importance within a cost group. For example, although the system C-13 shows a better benefit-cost ratio of 4:3 as compared to C-9 with the benefit-cost ratio of 4:1, the system C-9 in our judgment is more applicable to the chamber studies.

Also, it should be noted that the system C-1 is based on using only one instrument for the entire system. If two instruments have to be used for the smoke and the chemical agents studies, the benefit-to-cost ratio would be halved to 4.

#### 4. CONCLUSIONS

During this program, a survey of commercial aerosol instruments was conducted. Manufacturer's literature and published technical literature describing the principles, applications, and performance of these instruments were compiled.

Commercial aerosol instruments range widely in the degree of sophistication. Commercial mass monitors based on piezoelectric quartz microbalance have been sufficiently developed and automated for a variety of aerosol measurements.  $\beta$ -ray mass monitors are also considered adequate. The particle size analyzers based on the conventional optics are also automated and versatile but are being challenged by instruments based on laser optics. Most of the particle sizing is still based on light scattering methods. Other principles used for sizing are electrical mobility and automated cascade impactors.

Instruments specifically applicable to chamber smoke and chemical agents studies were chosen and their combinations were evaluated using a benefit-cost index. Following main conclusions were obtained:

Table 24  
BENEFIT/COST GROUPS

<u>System</u>	<u>Benefit/Cost</u>	<u>Cost (\$ ,000)</u>
C-1	8.1	12.0
C-13	4.3	20.8
C-9	4.1	26.1
C-12	3.7	24.9
C-11	3.6	24.2
C-7	3.6	30.2
C-8	3.6	30.2
C-6	2.0	57.2
C-5	1.9	46.7
C-3	1.8	50.8
C-4	1.8	50.8
C-10	1.8	51.9
C-2	1.2	77.8

1. The chamber aerosol concentration was too high for all of the commercial instruments. An auxiliary dilution system described in Section 3.1.5 may be used for obtaining dilution factors of up to  $1:10^3$ . Such a system is not commercially available at present. However, a satisfactory dilution system may be fabricated for under \$10,000 and may be used with any of the instruments described.
2. Commercial instruments are available for automated monitoring and sizing of the aerosols under study after dilution. The methods which form the basis of rapid real time monitoring are mostly based on light scattering principles or other in-flight sensing techniques. The measurements are subject to errors and biases associated with the measurement technique and their absolute accuracy cannot be compared with extractive techniques such as filter sampling and cascade impactors. It was, however, pointed out that the measurement technique employed should be comparable to the properties under study. Light scattering instruments are especially adapted to the smoke studies. The cascade impactor microbalance may be suited better for the solid agents but does not cover the complete range of desired requirements.
3. The benefit-cost index approach is useful to indicate useful combination instrument systems. Selection of instruments on a comparable rating of the benefit-to-cost ratio should be based on their performance and applicability of principle to the aerosol properties under study.

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